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PRELIMINARY DATA ON THE OCCURRENCE OF TRANSURANIUM NUCLIDES IN THE ENVIRONMENT AT THE RADIOACTIVE WASTE BURIAL SITE MAXEY FLATS, KENTUCKY

OFFICE OF RADIATION PROGRAMS, WASHINGTON, D. C.

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16. Abstracts

Between 1963 and 1974, approximately 104,000 cu/m of solid "low-level" radioactive waste were buried at the Maxey Flats, Kentucky, site. These wastes contained approximately 80 kg of plutonium-239 and a large undertermined quality of other plutonium isotopes. In 1972, elevated levels of radioactivity were detected in monitoring samples collected near the burial facility by the Kentucky Department for Human Resources (KDHR).

Subsequently, the KDHR conducted a special radiological study of the burial site and its environs. Based on the concentration of plutonium present or the ratio of plutonium-238 to -239, 49 of 50 samples collected on or near the burial site were contaminated with plutonium from a source other than atmospheric fallout. Plutonium was detected in surface soil, in soil cores 90 cm deep, in monitoring wells, and in streams which drain the site. This paper presents the plutonium data with interpretations of its occurrence and possible significance from a hydrogeological and environmental perspective.

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Water wells,
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Geologic structures,
Monitoring,
Fluid infiltration,
Runoff,
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U. S. ENVIRONMENTAL PROTECTION AGENCY
Office of Radiation Programs

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G. Lewis Meyer

February 1976

U.S. ENVIRONMENTAL PROTECTION AGENCY
OFFICE OF RADIATION PROGRAMS
WASHINGTON, D.C. 20460

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FOREWORD

The Office of Radiation Programs carries out a National Program designed to evaluate man's exposure to ionizing and nonionizing radiation and to promote development of controls necessary to protect the public health and safety and assure environmental quality.

Within the Office of Radiation Programs, problem areas have been defined and assigned a priority in order to determine the level of effort expended in each area. One of these, the waste management problem area, has been assigned a high priority and requires the participation and cooperation with several State and Federal agencies. This is one of a series of reports directed at a specific Environmental Protection Agency task of establishing action guidelines for radioactive waste management and disposal based on radiation exposure levels. Other reports, recommendations, and State assistance are being developed and executed to fulfill EPA obligations in the management and disposal of all types of radioactive waste including high-level wastes, low-level wastes, transuranium-contaminated wastes, mill tailings, naturally-occurring radioactive wastes and wastes from decommissioned nuclear facilities.

This report discusses the disposal of low-level wastes at a specific site, Maxey Flats, and the possibility of whether plutonium is migrating from the trenches. The important points in this paper are: (1) plutonium and other radionuclides are out of the trenches; (2) the data suggest that plutonium may be migrating through the ground; and (3) this occurrence can and must be prevented in the future.

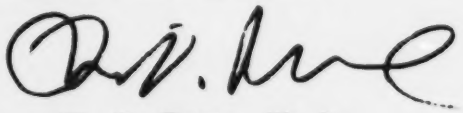
The potential for a health hazard is not an issue in this paper. Health authorities from the State of Kentucky, the Nuclear Regulatory Commission, and EPA have all advised that the levels of plutonium detected in the environment at Maxey Flats do not constitute a health hazard at this time. Likewise, EPA earth scientists do not believe that the mechanisms suggested in this paper regarding plutonium's mobility in the environment apply to the disposal of plutonium in a deep geologic disposal situation such as has been suggested for the ultimate disposal of high-level and transuranium-contaminated wastes. Rather, the problems discussed herein are believed to be associated with the disposal of plutonium and other radionuclides in shallow landfills in humid climates using present methods.

The Nuclear Engineering Company, Inc., under the direction of the Kentucky Department for Human Resources and with recommendations from the Nuclear Regulatory Commission, has taken significant actions which have entailed considerable expenditures in time, money, and manpower to correct the problems described herein. These efforts are recognized but were not within the scope of this report which basically is a scientific report using this site as a field laboratory to evaluate the existence of pathways for the movement of buried radioactive waste to the environment.

Review comments were received from the Nuclear Regulatory Commission (NRC), the Energy Research and Development Agency (ERDA), and the Nuclear Engineering Company, Inc. (NECO) and were quite valuable to the final editing of this report. Where it seemed appropriate, the suggested changes were incorporated into the report; in some cases EPA disagreed with the comment. Several valid suggestions were too cumbersome to incorporate within our publication schedule. The comments of ERDA and NRC and our responses to them are included as Appendixes D and E. More than one hundred comments and considerable exhibits were submitted by NECO. These comments were considered in detail and some suggested changes and data were incorporated; they are, however, too lengthy to present herein.

Five preliminary EPA-funded studies at the Maxey Flats facility are in the final stages of completion. These studies cover the hydrogeology, the occurrence of radionuclides, the pathways for radioactivity to man, and the inventories of wastes buried at the site. Publication of these reports has been given a high priority and is expected within the next few months.

I encourage users of this report to inform the Office of Radiation Programs of any omissions or errors. Your additional comments or requests for further information are also solicited.



W. D. Rowe, Ph. D.
Deputy Assistant Administrator
for Radiation Programs

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The author is grateful to the Kentucky Department for Human Resources for the use of the Pu data from their "6-month" report. David T. Clark was requested to participate as a co-author of this paper but could not because of pressing official duties; however, details about the site, its operations, and the sample data furnished by Mr. Clark were invaluable.

Other investigators at the site including H. Kolde (USEPA, evaporator study) D. M. Montgomery (USEPA, environmental pathways study) and H. H. Zehner (USGS, hydrogeological study) and the staff of the Nuclear Engineering Company, Inc. (Site operator) who generously gave access to preliminary data from their studies and contributed through stimulating discussions of the phenomena they observed and by critically reviewing this paper. Thanks are also due to I. J. Winograd and J. E. Dieckhoner for their critical reviews and discussions.

Special acknowledgement is given to Philip S. Berger, for his assistance in preparing this paper, and to Mrs. Hattie M. Leonard and Ms. Ella M. Thomas of the clerical staff for typing it.

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ABSTRACT

Between 963 and 1974, approximately 104,000 m^3 of solid "low-level" radioactive waste were buried at the Maxey Flats, Kentucky, site. These wastes contained approximately 80 kg of plutonium-239 and a large undetermined quantity of other plutonium isotopes. In 1972, elevated levels of radioactivity were detected in monitoring samples collected near the burial facility by the Kentucky Department for Human Resources (KDHR).

Subsequently, the KDHR conducted a special radiological study of the burial site and its environs. Based on the concentration of plutonium present or the ratio of plutonium-238 to -239, 49 of 50 samples collected on or near the burial site were contaminated with plutonium from a source other than atmospheric fallout. Plutonium was detected in surface soil, in soil cores 90 cm deep, in monitoring wells, and in streams which drain the site.

During the past 13 years, infiltrating precipitation collected in the burial trenches, forming a mobile plutonium-contaminated leachate. The chemical form, pathways, and mechanisms by which the plutonium moved out of the trenches have not been explained satisfactorily. Hydrogeological, radiological, and environmental pathways studies of the burial site are in progress.

The primary conclusion drawn from the hydrogeology of the site and the environmental radioactivity data is that plutonium has moved from the facility; possibly via several pathways including surface water runoff, atmospheric fallout from an evaporator, lateral migration through the soil, and migration through jointed subsurface geologic formations.

This paper presents the plutonium data with interpretations of its occurrence and possible significance from a hydrogeological and environmental perspective.

1. INTRODUCTION

1.1. Background

The State of Kentucky authorized the establishment of a commercial radioactive waste disposal facility in the State in early 1962 in the hopes of attracting nuclear industry [1]. A site in Fleming County known as Maxey Flats was evaluated and was deemed suitable for the disposal of solid radioactive waste. In January 1963 the Nuclear Engineering Company, Inc. (NECO) was issued a license to operate the disposal facility and in May 1963 the first radioactive material was buried at Maxey Flats.

In 1971, the Kentucky Department for Human Resources (KDHR) decided additional studies were needed at Maxey Flats because (1) of the exponential increase in quantities of radioactive materials, particularly transuranium-contaminated wastes, and (2) of growing concern that precipitation infiltrating the completed trenches and saturating some of the buried wastes might affect the containment of the radioactive material.

In late 1972 environmental monitoring detected elevated levels of radioactivity in the Maxey Flats environs. In November 1973 the KDHR initiated a six-month special environmental monitoring study [2] to identify the source and extent of these increased levels of environmental radioactivity. More than 50 samples were collected and analyzed for Pu-238 and Pu-239 as part of this study.

The KDHR in their six-month study and the independent Environmental Protection Agency (EPA) through indepentent investigations concluded that the disposal site at Maxey Flats was contributing radioactivity and Pu to the environment. As a result, the State limited further burial of Pu and other transuranium-contaminated wastes at Maxey Flats by license modification in 1974.

It should be emphasized that both State and EPA scientists have stated that the activity detected in the environment around Maxey Flats does not create a public health hazard at this time [2, 3]. For example, the levels of Pu detected in the samples collected during the KDHR's six-months study are only a few thousandths of the Maximum Permissable Concentration listed in the National Bureau of Standards "Handbook 69" and Kentucky Administrative Regulations 902 KAR 100: 025. The potential for health hazard is not an issue in this paper. The important points are that (1) Pu and other radionuclides are out of the trenches and (2) it appears that Pu may be migrating via the water pathways.

1.2. Scope

The hydrogeology of Maxey Flats, the Pu data, and an interpretation of their interrelation are presented from a straightforward hydrological point of view. The literature on Pu and its occurrence in soils and its mobility in the ground is also reviewed. The following quote exemplifies a widespread view of Pu in the ground:

> " Chemical and physical characteristics of plutonium (the principal transuranium element) are such that migration in soil or groundwater is unlikely. Deep well samples taken at the perimeter of the burial sites have not shown any detectable plutonium, thus, indicating that the buried plutonium has remained immobile." (From USAEC announcement of proposed rule-making on disposal of transuranium-contaminated wastes.) [4]

The occurrence of Pu at Maxey Flats, as seen from the data collected by the KDHR, raises some questions about our concepts of Pu mobility in the environment. For this reason, it seems important to present the data and a possible interpretation of its occurrence and migration to the experts on plutonium geochemistry. The four major objectives of this paper are: (1) to present the Pu data from Maxey Flats; (2) to determine if Pu is migrating from the burial site; (3) if Pu is migrating, to determine how and by what pathways migration may be occurring; and (4) to comment on the significance of the Pu data from a hydrogeological and environmental perspective.

2. BURIAL OPERATIONS

2.1. General Operations

Low-level radioactive wastes are shipped into the Maxey Flats burial facility by truck, and until 1972, both solid and liquid wastes were received for burial [1]. The receipt of liquid wastes was prohibited thereafter.

Most wastes are shipped to Maxey Flats in 55-gallon steel drums or in wooden and cardboard boxes and are buried in their shipping containers. The primary consideration in the design of the "burial" container is containing the wastes during shipment and protecting the workers. The containment capabilities of the steel drums under water- and leachate-saturated conditions are negligible, especially since many are crushed when dumped randomly into the trench and during compaction of the trench cap.

With few exceptions, the solid wastes are buried in large rectangular common trenches and covered with earthen caps. These trenches range in length from 76 to 110 m and re 15 m wide and 6 m deep. Trenches are excavated to full size before burial begins. The floor of the trench is sloped one degree to a sump and a stand pipe is placed in the sump. After being logged in and surveyed, the truck with the wastes is backed to the working face of the trench and the wastes are dumped randomly into the trenches from the truck. Particularly large items and those items emitting high levels of radiation in casks are emplaced by crane. In the past, the wastes were covered with dirt daily to reduce radiation exposure and spread of contamination. Now, they are covered temporarily with a reinforced plastic cover.

When filled, the trench is capped with a minimum of 1 m of the soil and shale excavated from it. The cap is mounded to encourage runoff and is compacted with a sheepsfoot roller to reduce infiltrating precipitation and facilitate runoff. Since 1971 shallow-rooted ground cover, such as fescue grass and clover, has been planted to prevent erosion.

Liquid wastes received from off-site in the past were solidified on site and placed in special "L" series trenches. Commonly, the liquids were mixed with cement and paper in a cement mixer and the cement-paper mixture was poured into a polyethylene-lined trench. When the trench was filled, the polyethylene sheeting was lapped over the top of the radioactive cement-paper mixture and the trench was capped with earth. Although disposal of off-site liquids was stopped in 1972, large volumes of radioactive liquid wastes were until recently being generated onsite in the trenches by infiltrating precipitation. The treatment of these liquid wastes is described later in Section 6.8.

2.2. Water Management Program

By 1972, some of the completed trenches were filled or partially filled with infiltrating precipitation. A water management program was initiated to remove this water from the trenches and to prevent further entry of water. The two-part program was designed on the assumption that most of the water in the trenches originated from precipitation falling onto the trenches rather than from groundwater. One part was directed towards water removal and included: (1) dewatering the trenches by pumping; (2) storing the leachates in surface storage tanks; (3) reducing the volume of the leachate by evaporation; (4) solidifying the evaporator residues; and (5) burying the solidified residues on-site. The other part was directed at reducing the infiltration of precipitation and included: (1) adding fill to the caps; (2) compacting the caps further; (3) reshaping the cap profiles to encourage runoff; (4) designing and engineering an integrated surface drainage system for the site; and (5) planting and maintaining shallow-rooted ground cover to reduce erosion of the cap.

There is no question that the water management program has had many beneficial effects in reducing the potential for the migration of radioactive contaminants from the trenches. It has not been without minor problems, however. Dewatering and evaporation operations have reduced leachate accumulation in the trenches; however, spillage of the leachate during pumping operations and leakage from the storage tanks have caused some surface contamination [6]*. The evaporator plume may have added another potential pathway for the migration of Pu; however, there is no reason to believe that the consequences of this pathway are large. The cap renovation program has halted erosion of the caps and should reduce the infiltration of precipitation into the trenches. However, growth of a shallow-rooted ground cover on the caps may, to some degree, maintain a degree of permeability in them.

As of yet unpublished data from KDHR and EPA studies [8, 9, 10] indicate that water management activities (trench dewatering and leachate evaporation) have been a source of small but measurable surface contamination which at present does not pose an immediate health hazard. However, this low-level of contamination, together with the hydrogeology of the site, greatly complicates scientific studies at the site. For example, several natural paths for surface runoff and overland flow cross over several potential points of subsurface discharge, obscurring, or at least making it difficult to determine whether the source of contamination detected is from subsurface discharge or from operational activities.

Viewed in the long term, it is difficult to imagine that either the State or the operator will continue to dewater the trenches for hundreds or thousands of years---the hazardous lifetime of the wastes. Yet, unless the cap improvement program is truly effective in developing a nearly impervious cap, long-term pumping could be required.

* NECO reported that all leakage from the tanks was safely contained within a berm around the tank farm with no hazard to operating personnel or the public [7].

3. SITE DESCRIPTION

3.1. General Setting

The burial ground is situated on Maxey Flats, a flat-topped, highly-dissected ridge in the Knobs region of Kentucky. The ridge rises approximately 100 m above the wide, flat, alluvial-filled valleys (locally called hollows). The upland surface is gently rolling and is generally less than 600 m wide.

The Maxey Flats region has a humid continental climate with sharp contrasts between winter and summer temperatures. The mean annual precipation ranges from 100-120 cm and averages about 110 cm [11]. Much of the region is cleared for agricultural use; tobacco, corn, and forage are the major crops. A few scattered farm families live along the ridge and in the hollows; however, the area around Maxey Flats has a low population density, in general less than 4-10 people per km^2 [12]. Only the upland surfaces and slopes are heavily forested. These are predominantly the oak-hickory ecotype and some may be uncut, first growth forest. The chief mineral resources are limestone, sand, and gravel.

3.2. Geology

Maxey Flats is located on the eastern flank of the Cincinnati arch and is directly underlain by shales, siltstones and sandstones that gently dip to the southeast (4.7 m/km). A generalized geologic cross-section of the rocks that outcrop at the burial site and a brief lithologic description are presented in Figures 1 and 2 [13].

At Maxey Flats only the lower 12 m of the Nancy Member of the Borden Formation are present. The trenches are entirely within the Nancy Member. It is a poorly fissile, green shale, which is plastic when wet and which has siltstone and sandstone interbeds. The Farmers Member, the lower unit of the Borden Formation, directly underlies the Nancy Member. It is a ledge-forming, well sorted, indurated, very fine grained, evenly bedded, quartzose sandstone with shale interbeds less than 1 m thick. This highly competent formation has well developed jointing and fracturing. The Henley Bed, a 1.5 m thick, greenish-gray clayey shale with 2-5 cm thick sandstone and siltstone interbeds, lies at the base of the Farmers Member.

Papadopulos and Winograd [14] wrote that open joints and bedding planes can be seen in outcrops of the sandstones and siltstones at the base of the Nancy Member in the Farmers Member and Walker [15]

FIGURE 1. GENERALIZED GEOLOGICAL CROSS-SECTION OF THE MAXEY FLATS REGION, KENTUCKY.

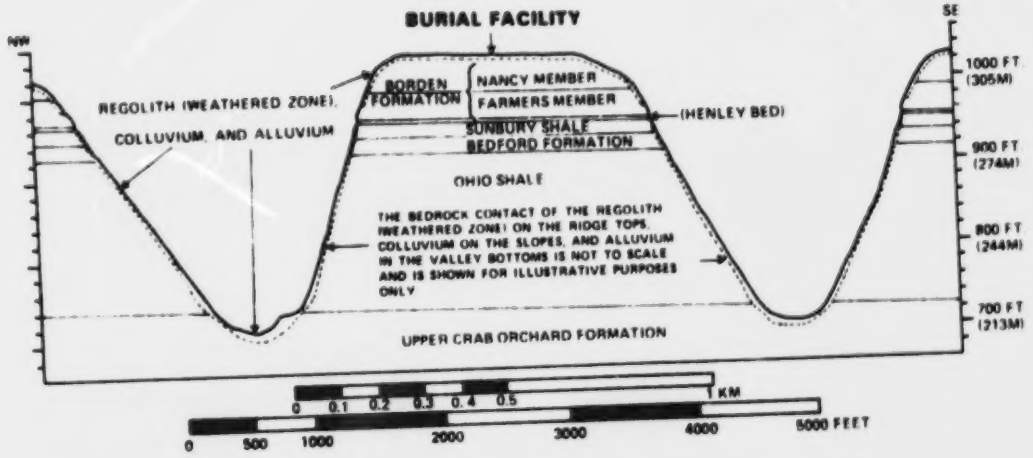
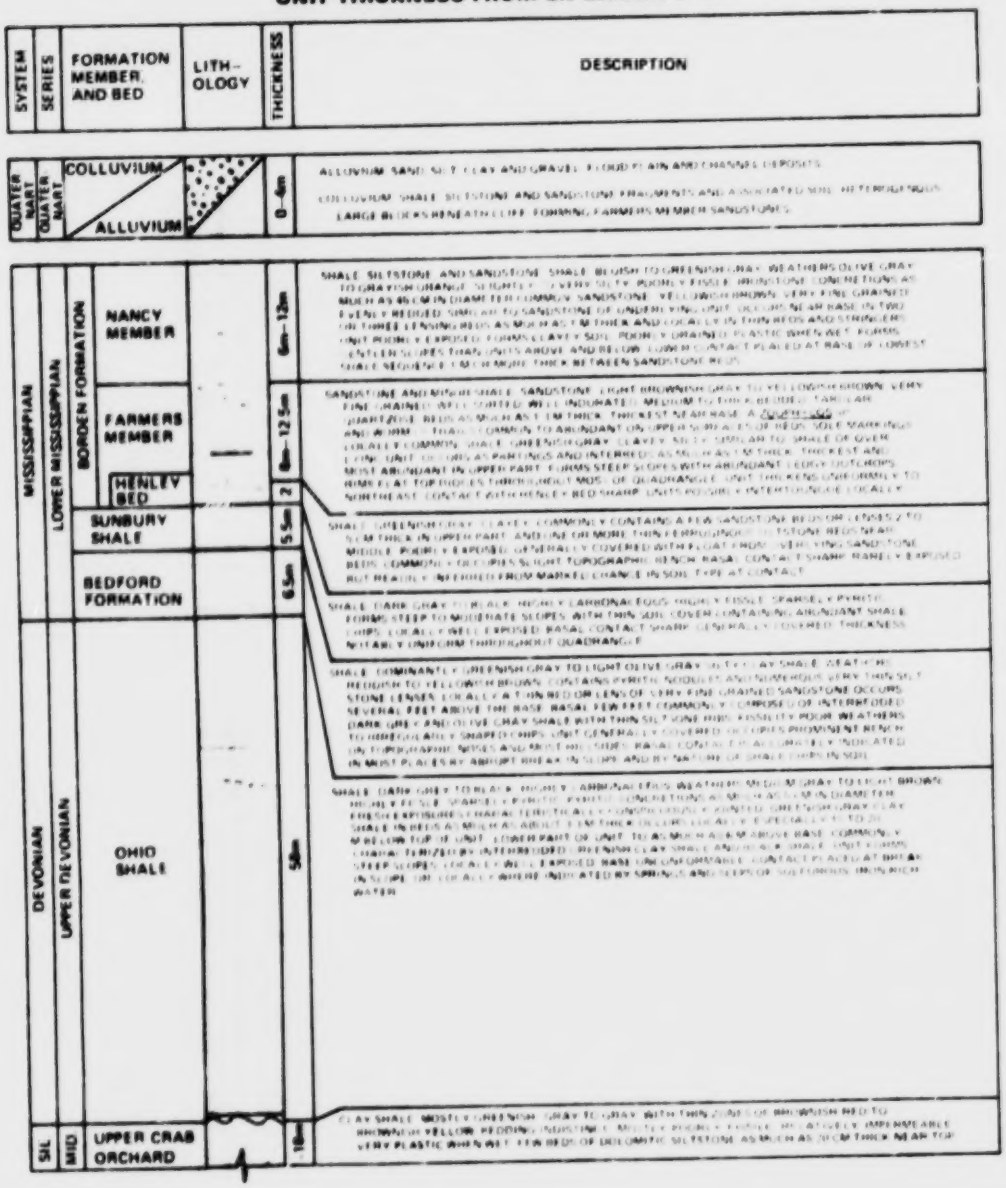

BURIAL FACILITY
NW
SE
REGOLITH (WEATHERED ZONE), COLLUVIUM, AND ALLUVIUM
BORDEN FORMATION
NANCY MEMBER
FARMERS MEMBER
(HENLEY BED)
SUNBURY SHALE
BEDFORD FORMATION
OHIO SHALE
THE BEDROCK CONTACT OF THE REGOLITH (WEATHERED ZONE) ON THE RIDGE TOPS, COLLUVIUM ON THE SLOPES, AND ALLUVIUM IN THE VALLEY BOTTOMS IS NOT TO SCALE AND IS SHOWN FOR ILLUSTRATIVE PURPOSES ONLY
UPPER CRAB ORCHARD FORMATION
1000 FT (305M)
900 FT (274M)
800 FT (244M)
700 FT (213M)
0 0.1 0.2 0.3 0.4 0.5 1 KM
0 500 1000 2000 3000 4000 5000 FEET

FIGURE 2. STRATIGRAPHIC COLUMN AND LITHOLOGIC DESCRIPTION OF THE GEOLOGY AT MAXEY FLATS, KENTUCKY (AFTER MCDOWELL, ET AL.) UNIT THICKNESS FROM EN EMCON (17).



noted the occurrence of small quantities of water seeping from well-developed joints in the lower part of the Borden Group (now the Borden Formation).

3.3. Hydrology

Papadopulos and Winograd [14] have recently described the occurrence of groundwater at Maxey Flats. The uppermost water table is perched in the soil zone above the poorly permeable Nancy Member and slopes southeastward paralleling the regional dip. This water table supplies the shallow dug wells on Maxey Flats. Hopkins [16] reported that deeper wells elsewhere on Maxey Flats have water levels as much as 9 m below the perched water in the soil zone. The lower water levels may represent a lower perched water table or may be evidence of decreasing head with depth, which would suggest vertical ground water movement, probably through joints and fractures, within the Nancy Member. Other perched water tables may exist within the sandstones of the Farmers Member and conceivably in the Sunbury Shale above the reportedly expansive clay-shales of the Bedford Formation [17].

The main water table occurs in the Ohio Shale [17]. This unit supplies several domestic wells in the Maxey Flats region. The Upper Crab Orchard Formation, a shale which is plastic when wet, underlies the Ohio Shale and may be thought of as the hydrologic basement for the Maxey Flats site. Papadopulos and Winograd [14] point out that the near-perennial nature of the creeks in the hollows surrounding Maxey Flats indicates that the creek discharge is primary base flow from the Ohio Shale and/or the Upper Crab Orchard Formation and possibly from the colluvium blanketing parts of the slopes around the site.

The formations at Maxey Flats are, in general, aquitards through which the intergranular movement of groundwater is very slow. However, hydraulic tests of wells indicate that water can move along the joints and bedding planes. Walker [15] reported that the loss of water during a pressure test of a well* indicated that there are individual channels (joints) large and continous enough to constitute a potential pathway for the migration of contaminants from the trenches. EMCON [18] reported a 100 percent loss of drilling water a number of times in the Nancy, Farmers, Sunbury, and Ohio Shale during drilling and coring operations. More recently, Zehner [17] reported conducting withdrawal hydraulic tests which indicate that water movement occurs along joints and/or bedding planes.

* Assuming that no leakage occurred around the packers, the results of Walker's tests indicate that injection rates range from 0.00006-0.015 [(1/s)/m] (kgf/cm^3) [14].

It should be emphasized that the formations at Maxey Flats are aquitards with very low effective porosity. However, this low effective porosity, which occurs along joints and bedding planes, could cause the channeling and movement of groundwater at unexpectedly high velocities.

3.4. Hydrogeology and Potential Pathways

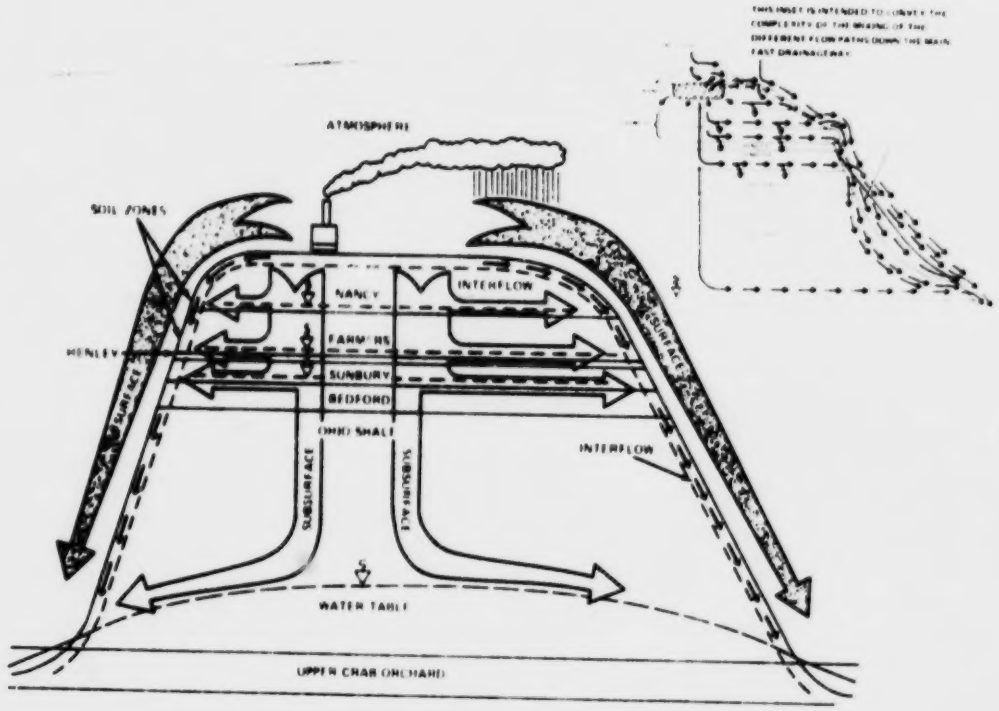
Papadopulos and Winograd [14] summarized the present knowledge about the hydrogeology of Maxey Flats as follows, "...the stratigraphy of Maxey Flats is well known and its local hydrologic boundaries approximately defined" but, "the hydrogeology of the site is poorly understood." They further warn that, "The strata beneath Maxey Flats...are aquitards whose fractured nature and low permeability constitute major obstacles to successful quantitative definition of the hydrogeology of the region, regardless of funding." It is hoped that the preliminary hydrogeologic investigation presently underway [19] and the more detailed investigation soon to be commenced [20] will show them to have been pessimistic.

The basic hydrogeologic (and water pathways) model for Maxey Flats is complex. It is believed that water is the primary mobilizing agent for Pu, that it is the primary driving force for the Pu, and that it is the primary pathway from the burial site at this time.

Almost all water discharging from the site and from the subsurface formations beneath it originates from precipitation falling on the site. This water discharges from the site by the following paths: surface runoff; interflow or movement through the shallow soil zones; and subsurface bedrock flow. These three flow paths form the three potential water pathways for the migration of Pu from the site. The evaporator plume may constitute a fourth pathway for Pu. Figure 3 represents a schematic of the major flowpaths for water (pathways for contamination) passing through the system.

Water entering the ground as recharge moves downward and laterally through the hydrogeologic system with its movement complicated by differences in permeability between strata, the dip of the strata, and extensive jointing. There is strong theoretical evidence for this interpretation as well as numerous examples from like hydrogeolgic settings. Walker [15], Whitman [21], Papadopulos and Winograd [14] and Zehner [19] also used this basic interpretation. Field evidence, the presence of perched water beneath the site, and repeatedly measured water levels in many of the E-Series wells [17] further supports this interpretation.

Figure 3. Schematic diagram of water pathways at Maxey Flats



It is believed that the gentle dip of the strata to the east-southeast at 4.7 m/km causes much of the perched and main water tables to slope and discharge to the southeast. A groundwater divide certainly must exist beneath the burial facility because water discharges all along the flanks of Maxey Flats beneath the burial facility. Its location may be, however, altered by the infiltration of leachates from the trenches. The dip of the strata, plus grading and earthmoving activities also cause the land surface at Maxey Flats to slope southeasterly, channeling most surface runoff from the site into a main east drainageway.

The extensive jointing throughout the Nancy, the Farmers, the Sunbury, and the Ohio Shale probably causes the greatest complications. Whereas the layering and slope just described tend to channel and direct the movement of water through the system with some predictability, the fracturing makes it very difficult to predict the direction and rate of subsurface flow. It also greatly increases the permeability of a series of otherwise virtually impermeable shale, siltstone, and sandstone aquitards.

A less obvious but equally serious effect of jointing and fracturing is the reduction in the ion exchange reaction between the rock media and contaminants. Most of the strata are shale and siltstone, which usually have good-to-excellent ion exchange capacities when pulverized and tested in the laboratory. The ion exchange interaction between the earth media and contaminated liquid (leachate) is usually counted on as an important safety factor in the land burial of radioactive wastes. This presupposes that contaminants dissolved from the wastes will come in contact with and be sorbed on the clay particles during their migration by intergranular flow through the formations. If the contaminants migrate along fractures as probable at Maxey Flats rather than between individual silt- and clay-size particles, the effective surface area of the silt and clay exposed to the contaminant is greatly reduced, thus by-passing the ion exchange process to a large extent.

Logs of test wells drilled on Maxey Flats commonly report the presence of vertical joints or fractures. Many of the joints have been described as weathered or iron-stained [18], indicating that groundwater has passed through the joints. Iron-stained joints can also be seen in the walls of the trenches excavated into the Nancy Member and in outcrop. The jointed character of the burial media, the Nancy Member, can be seen in Figure 4. The underlying Farmers Member, Sunbury Shale and Ohio Shale Formations are also highly jointed. [13, 15, 14, 18]

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Figure 4. Photograph of jointed Nancy Member (picture was taken while standing in bottom of an open trench).

4. SOURCE TERM

4.1. Waste Volume and Activity

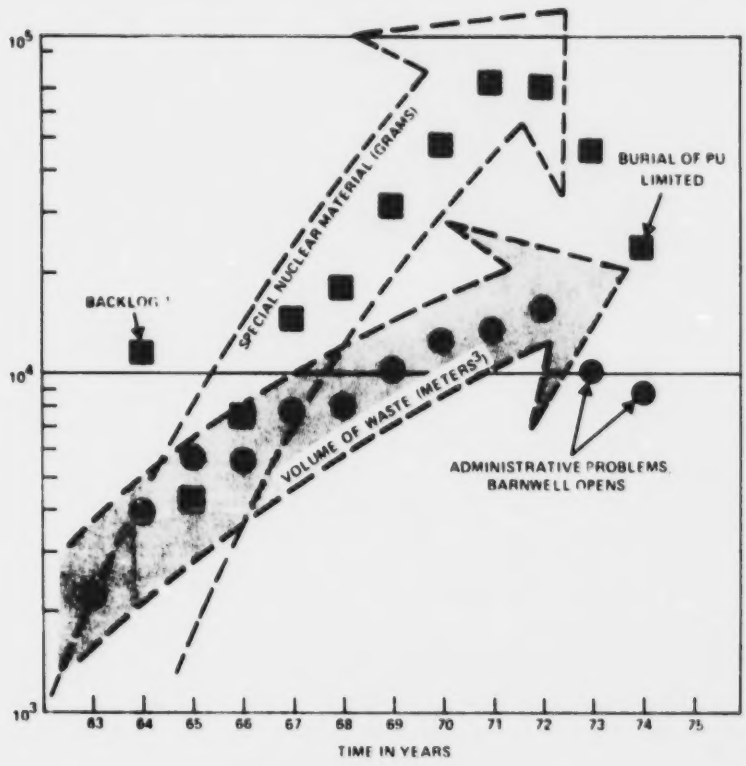
From 1963 when disposal operations began through 1974, 104,000 cubic meters of solid and 2,250,000 l of solidified liquid radioactive wastes were buried at Maxey Flats. These wastes included 1,638,000 curies of by-product materials *, 349,000 grams of special nuclear material (SNM)*, and 158,000 pounds of source material* [1]. The volume of of solid wastes buried at Maxey Flats represents 35% of the volume and 65% of the activity of all wastes buried at commercial burial facilities in the United States during 1963-1974.

It became clear quite early that the assumed burial rate of 750 curies per year was a gross underestimation [1]. The actual and projected (as of 1972) annual rates of burial are shown in Figure 5. The annual volume of waste buried at Maxey Flats increased almost exponentially through 1971. The apparent decrease in burial rate in 1971-1972 is believed to have resulted from labor/administrative difficulties at Maxey Flats and to diversion of waste to the commercial disposal facility at Barnwell, South Carolina, which opened in 1971.

Of direct interest to this paper is the increase in the amount of special nuclear material being buried between 1963 and 1972 --a doubling approximately every 1.3 years (Figure 5). The State limited further burial of Pu and other transuranium-contaminated wastes at Maxey Flats by license modification in 1974 partly it is believed as a result of this phenomenal growth in the burial rate of special nuclear material. The distribution and quantities of Pu-238, Pu-239, and other specific isotopes in the SNM category which were buried at Maxey Flats are not known. Although the Pu-239 was recorded separately and more than 80 kg were reportedly received, the accuracy of the records is uncertain [1]. Preliminary printouts of data from a detailed inventory indicate that many shipments were recorded only as Pu, Pu-xxx, U, and U-xxx [22]. Better identification of transuranium isotopes buried at Maxey Flats may require reviewing individual invoices with the originators of the waste.

* By-product material (radioisotopes produced in reactors), special nuclear material (plutonium and enriched uranium), and source material (uranium and thorium) are defined in USAEC Rules and Regulations, 10 CFR 20 [23].

FIGURE 5. PROJECTIONS OF BURIAL GROWTH RATES FOR VOLUMES OF WASTE AND QUANTITIES OF SPECIAL NUCLEAR MATERIAL AT MAXEY FLATS BURIAL FACILITY (AS OF 1972).



In a preliminary inventory [1], detailed hand tabulations were made from the shipping records of two trenches. In Trench 30, 2,300 g of Pu-239 were identified out of a total of 48,000 g SNM. In Trench 32, 13,000 g of Pu-239 were identified out of a total of 54,000 g SNM.

4.2. Waste Character

"Low-Level" radioactive waste is now called "other than high level waste" and now includes everything except "high-level" radioactive waste*. In brief, any waste, including transuranium contaminated wastes, which is not "high-level" waste can now be buried at commercial disposal sites, regardless of its activity or hazard potential, unless there are specific site regulations against it.

Most of the wastes buried at Maxey Flats are believed to be large volume, low-hazard-potential solid wastes such as paper trash, cleanup materials and liquids, packing material, broken glassware, plastics, protective clothing, radioactive carcasses of experimental animals and contaminated equipment. The most recent description which the author could find of the wastes actually being buried at commercial burial sites [24] estimated that 70 percent by volume of these wastes would be paper materials and that the density of the total waste would be about 10 lbs/ft (0.16 gm/cm^3). High-activity wastes such as sealed sources, reactor resins, filters, and irradiated reactor parts have also been buried. A large number of records list the isotopes shipped only as "mixed fission products," "low specific activity," "not specifically identified," and other non-specific identifications [1].

KDHR and EPA studies [1, 22, 25] have found that for practical purposes no information is available on the chemical or physical character of the wastes which have been buried at Maxey Flats.

* The AEC divides radioactive waste products into two categories, "high-level wastes" and "other than high-level wastes". "High-level wastes are defined as, "aqueous waste resulting from the operation of the first cycle solvent extraction system, or equivalent, and the concentrated waste of subsequent extraction cycles, or equivalent, in a facility for reprocessing irradiated reactor fuels". The AEC group all other wastes in the category, "other than high-level wastes" [23].

5. RADIOCHEMICAL DATA

5.1 Data from "6-Month" Study

The scope of this paper has been limited to a detailed examination and interpretation of the Pu data collected by the KDHR in a special 6-month study of the burial site and its environs, 1973-1974. Literally, thousands of additonal analyses are available for other isotopes from other studies and which contained H-3, Co-60, Sr-89, Cs-134, and Cs-137, in concentrations greater than background or fallout, indicating that other radioactive contaminants have moved or migrated also; thus tending to support the conclusion that Pu has moved or migrated. These additional data have been used or referenced occasionally to confirm conclusions reached from (1) the Pu data and (2) knowledge of the hydrogeology of the site. However, extensive discussion of these additional data was not possible in this report. There are exceptions, however, which include (1) approximately 40 Pu analyses of leachate which was stored in the tank farm, (2) additional Pu analyses of samples from wells, and (3) listings of other radionuclides detected in effluents from Maxey Flats.

Four reports are presently in preparation which will present to the interested reader the bulk of the additional data and a much broader picture of the environmental impact of burial operations at Maxey Flats. These include (1) the radiological impact of burial operations [10], (2) the impact of the evaporator as a potential pathway [8], (3) the pathways to man for radiocontaminants from the site [9], and (4) a detailed analysis of the radioactive waste inventory buried at Maxey Flats [22, 25].

The "6-month" study was conducted by the KDHR between November 1973 and May 1974 and covered a six-mile radius of the site [2]. During this study, more than 400 soil, surface water, and ground water samples were collected in and around the burial facility, including a number of control samples for determining background levels. All samples were analyzed for tritium, gross alpha, and gross beta. In addition, 36 samples were analyzed for specific gamma emitters and Sr-89 and Sr-90; approximately 60 samples were analyzed for Pu-238 and Pu-239; and 14 samples were analyzed for U-234 and U-238.

Results of the Pu analyses are presented in Table I. The analyses were performed by the USAEC's Health Services Laboratory (HSL), Idaho Falls, Idaho, and Environmental Analysis Division, LFE Laboratory (LFE), Richmond, California, using the analytical techniques described by Sill, Puphal, and Hindman [25] and the HASL Procedures Manual [26], respectively.

TABLE Σ. CONCENTRATION OF ^{238}Pu AND ^{239}Pu AND RATIO OF ^{238}Pu TO ^{239}Pu IN SAMPLES COLLECTED FROM SOIL, TEST WELLS, AND STREAMS IN AND AROUND THE MAXEY FLATS BURIAL FACILITY DURING THE KDHR'S "6-MONTH" STUDY.

	Sample ID	238 Pu	239 Pu	238/239 Pu	Sample (1) Location and (2) Description
Soil Samples	129	0.07	0.05	1.4	(1) Approximately 1.5 m from east end of Trench 10, (2) Soil core, 60-75 cm depth.
	129	0.11	0.14	0.8	(1) Approximately 1.5 m from northeast end of Trench 10, (2) Soil core, 0-50 cm depth.
	[illegible]	[illegible]	[illegible]	4.0	(1) Approximately 1.5 m from northeast end of Trench 7, (2) Soil core, 25-50 cm depth.
	131	0.13	0.06	2.2	(1) Approximately 1.5 m from northeast end of Trench 7, (2) Soil core, 50-75 cm depth.
	132	0.13	0.04	3.3	(1) Approximately 1.5 m from northeast end of Trench 7, (2) Soil core, 35-40 cm depth.
	133	0.04	0.02	2.0	(1) Approximately 1.5 m from east (low) end of Trench 7, (2) Soil core, 35-60 cm depth.
	134	8.1	1.9	4.3	(1) East end of Trench 26 north of trench complex 1.5 m, (2) Soil core, 50-75 cm depth.
	8	1.1	0.2	5.5	(1) Same core hole as sample 139, (2) Soil core, 25-50 cm depth.
	8	2.6	0.58	4.5	(1) same core hole as sample 139, (2) Soil core, 50-75 cm depth.
	64	0.22	0.1	2.2	(1) East end of Trench 26 (different core hole than samples 139, 140, and 141), (2) Soil core, 50-75 cm depth.
	0	0.1	0.1	1.0	west end of Trench 28, 1 m from fence, (2) Soil core, 50-75 cm depth.
	6	0.6	0.22	2.7	(1) Drainage ditch north of Trench 28, (2) Soil core, 0-25 cm depth.
	7	0.67	0.167	4.0	(1) East end of Trench 25, 1.5 m from end of trench, (2) Soil core, 75-92 cm depth.
	5	0.6	0.111	5.4	(1) East end of Trench 25, 1.5 m from end of trench, (2) Soil core, 0-25 cm depth.
	150	0.15	0.04	3.8	(1) East end of Trench 2, 1.5 m from end of trench, (2) Soil core, 0-25 cm depth.
Test Well Samples	160	7.5	0.22	34.1	(1) well 4E, total depth 15 m, (2) Suspended solids and sediments.
	161	0.6	0.12	5.0	(1) well 3E, total depth 29 m, (2) Suspended solids and sediments.
	162	33	0.54	59.4	(1) well 1D, total depth 34 m, (2) Suspended solids and sediments.
	2782	4.1	0.14	32.6	(1) well 4C, total depth 15 m, (2) Suspended solids and sediments.
	2785	22	0.47	46.8	(1) well 1D, total depth 34 m, (2) Suspended solids and sediments.
Off Site Samples	210	0.57	0.071	8	(1) Stream discharging from Drip Spring Hollow, (2) Silt, sand, and gravel from stream bottom.
	211	0.074	0.14	0.54	(1) Spring fed pond at McRoberts farm below Ohio Shale-Upper Crab Orchard contact, (2) Silt, sand, and gravel from stream bottom.
	214	1.42	0.3	4.7	(1) Spring apparently issuing from outcrop of Ohio Shale in draw leading to eastern main drainage from site, (2) Silt and clay at base of seepage.
	217	24	0.46	52	(1) Stream fed by same spring as No. 116 at road, (2) Clay and silt.
	218	0.13	0.26	0.49	(1) Sulfur spring near base of Maxey Flats at east side of entrance to No Name Hollow, (2) Silt, clay, and organic matter.
	219	0.034	0.048	0.69	(1) Pond in alluvium at McRoberts farm, Drip Spring Hollow, (2) Silt and clay.
	134	0.15	0.02	19.5	(1) west of well 12E, 73 m linear, 12 m elevation drop, not on main west drainage path; (2) Soil core, 0-15 cm depth collected 60 cm back into hill under shale out crop.
	135	0.12	0.02	12.0	(1) west of well 12E, 137 m linear, 30 m elevation drop at wet weather spring near shale outcrop, (2) Soil core, 0-25 cm depth.
	147	2.52	0.26	19.8	(1) Drainageway from west side of site to Sample Station 114, (2) Forest loam core, 0-25 cm depth.
On Site Samples	125	0.58	0.1	5.5	(1) At fence, 4 m north of main east drain, (2) Soil core, 0-25 cm depth.
	126	2.92	0.27	10.8	(1) At fence, 8 m south of main east drain, (2) Soil core, 0-25 cm depth.
	127	0.16	2.15	0.6	(1) Main east drain at fence, (2) Soil core, 0-25 cm depth.
	128	2.78	0.87	3.2	(1) Main east drain, 6 m above tractor road, (2) Soil core, 0-15 cm depth, immediately overlying sandstone layer.
	124	2.89	0.04	72.3	(1) Main east drain settling basin, just below fence, (2) Sediment core, 0-15 cm depth.
	125	0.09	0.25	0.4	(1) Main east drain at precipice formed by Farmers sandstone bed, (2) Soil core 0-25 cm depth.
	126	0.85	0.27	3.1	(1) North of main drain in vicinity of well 1E, (2) Forest loam core, 0-25 cm depth.
	130	0.18	0.01	18.0	(1) At Trench 27, in direction of well 10E, 15 m west of fence, (2) Soil core, 0-25 cm depth.
	137	0.12	0.03	4.0	(1) In drainage ditch at northeast corner of Trench 33L, (2) Soil core, 0-25 cm depth.
	138	2.49	1.68	1.7	(1) At northwest corner of tank farm, 8 m west of fence, (2) Soil core, 0-25 cm depth.
	142	0.64	0.03	21.3	(1) Main drainage at south gate, (2) Forest loam core, 0-15 cm depth.
	143	41.98	2.76	15.2	(1) South main drainage, 8 m elevation drop from south fence, (2) Soil core, 0-25 cm depth.
	144	0.19	0.50	38.1	(1) Main south drainage at precipice, near outcrop of Farmers sandstone, (2) Forest loam core, 0-15 cm depth.
	145	2.55	1.37	1.9	(1) Northeast site drain flowing west to east just inside berm, (2) Soil core, 0-75 cm depth.
	147	12.90	1.28	10.2	(1) Northeast site drain flowing west to east just inside fence, (2) Soil core, 0-25 cm depth.
	150	0.109	0.10	6.1	(1) Northeast end of tank farm, just inside berm, (2) Soil core, 0-75 cm depth.
	151	0.029	0.024	1.2	(1) Soil from new, unused trench No. 42, (2) Soil core, 0-25 cm depth.
	153	0.65	0.8	0.11	(1) east to east drain, southwest of Trench 2, (2) Soil core, 0-75 cm depth.
	154	0.15	0.07	2.6	(1) West of Trench 31, 3 m inside fence, (2) Soil core, 0-25 cm depth.
	157	28.6	0.09	18.2	(1) Southwest corner of evaporator building at east wing of concrete pad, (2) Soil saturated Pu loss from DISCHARGE STACK for evaporator.
	158	1.76	0.15	11.7	(1) Drainage ditch along west side of Trench 40, by sump pipe, (2) Soil core, 0-25 cm depth.

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Inter-laboratory comparison of Pu analyses for the "6-month" samples are not available. However, there is no question about the presence of Pu from a source other than fallout in the environment in and around the burial site. Both HSL and LFE detected Pu in concentrations well above background levels in almost all samples they analyzed. In addition, above background concentrations of Pu were also found in samples collected and analyzed by EPA's RNEB [3], Radiation Management Laboratories [7], and Teledyne Isotopes, Inc. [7], which supports the presence of elevated levels of Pu reported by HSL and LFE. When the data are placed in ascending order of concentration or ratio of Pu-238 and Pu-239 as in Figure 6a, 6b, and 6c, the analyses from the two labs appear from simple inspection to be normally distributed throughout the 50 samples. Therefore, from these simple tests, it has been assumed that (1) the Pu concentrations reported were real and (2) the concentrations reported for each sample were not significantly affected or skewed by the laboratory which did the analysis.

The data have been grouped into four suites for interpretation: (1) off-site samples; (2) on-site samples; (3) core samples in the trench area; and (4) well samples.

5.2 Sampling Methods

The samples collected for Pu analysis during the "6-month" study were composed of soils and fill material from the trench and operational areas; soil and organic material from the area surrounding the site; clay, silt, and sand from the stream beds, ponds, and spring outfalls; and suspended solids and sediments from the monitoring wells. No water samples were analyzed for the "6-months" study.

The soil samples were collected with either an Oakfield hand coring device or a "Little Beaver" power auger. The Oakfield coring device is pushed into the ground by hand and will take a core 25 cm long by 2.5 cm in diameter. With the use of extensions, cores can be taken to a depth of 90-100 cm, if no cobbles or hard rock are encountered. The "Little Beaver" is a gasoline-powered Archimedes screw-type auger which can, under favorable conditions, take a 10 cm-diameter sample to a depth of approximately 150 cm. Pond, stream, and spring outfall samples were collected by scooping up the upper 3-5 cm of the bottom material.

Samples of water and entrained solids from the wells were collected with a bailer with a foot valve. However, only the suspended solids from these samples were analyzed from the "6-months" study

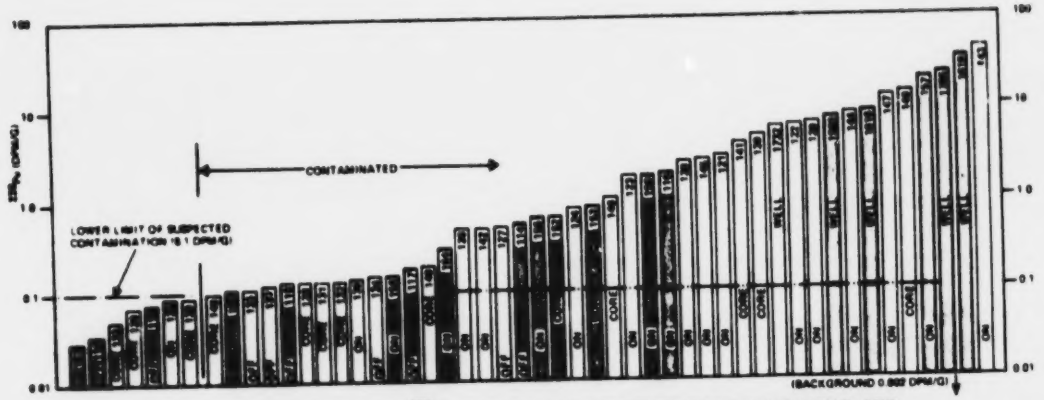


FIGURE 9A. CONCENTRATION OF ^{239}PU IN SAMPLES COLLECTED FROM SOILS, TEST WELLS, AND STREAMS IN AND AROUND THE MAXEY FLATS BURIAL FACILITY DURING THE KDHR'S "6-MONTH" STUDY.

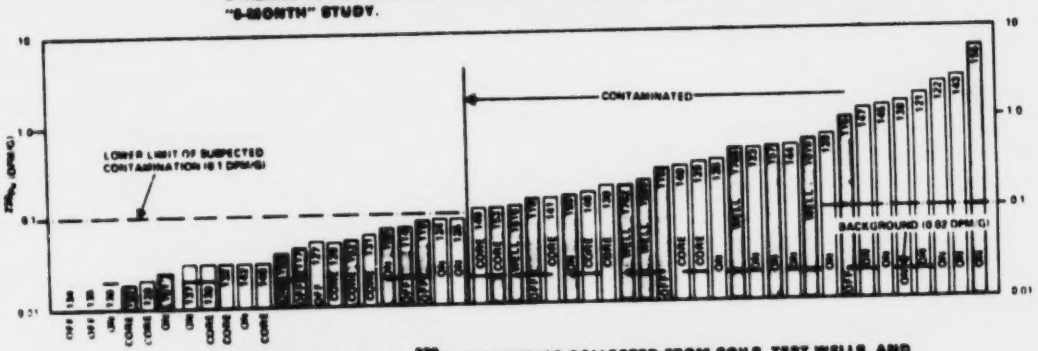


FIGURE 9B. CONCENTRATION OF ^{239}PU IN SAMPLES COLLECTED FROM SOILS, TEST WELLS, AND STREAMS IN AND AROUND THE MAXEY FLATS BURIAL FACILITY DURING THE KDHR'S "6-MONTH" STUDY.

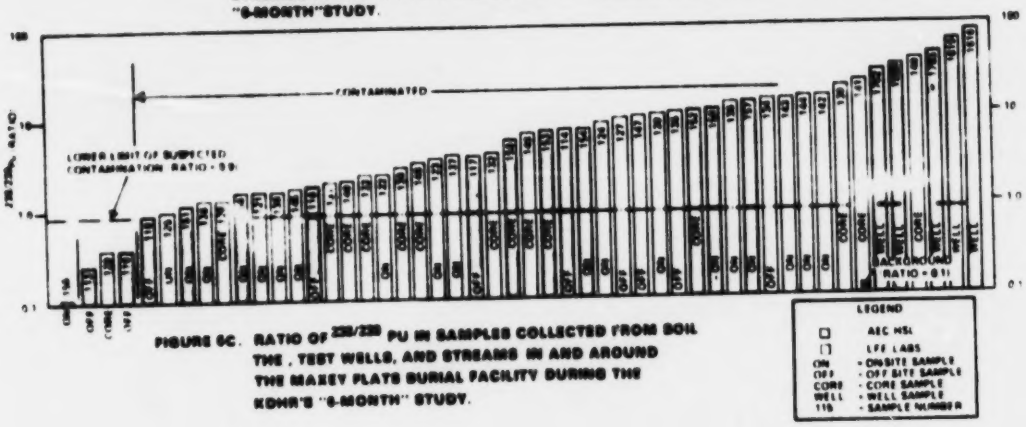


FIGURE 9C. RATIO OF $^{238/239}$PU IN SAMPLES COLLECTED FROM SOIL THE , TEST WELLS, AND STREAMS IN AND AROUND THE MAXEY FLATS BURIAL FACILITY DURING THE KDHR'S "6-MONTH" STUDY.

samples. Analyses are, however, available from the other studies [7, 9] for both suspended solids and the water from the wells. There is no way of knowing at what depth interval a sample was collected with this device.

The suspended solids in the well samples were filtered in the laboratory, ashed at 450 C, pulverized in a mortar, and passed through a 28-mesh sieve. From the portion that passed through, 5-10 g was removed, sealed in a plastic "Whirlpak" bag, and shipped for analysis. The other samples were dried in an oven, pulverized, and treated much like the well samples thereafter.

The question that must be asked at the outset is whether the "6-month" samples were inadvertently contaminated during the collection process by using contaminated samples devices. And if this was a possibility, would the samples therefrom be totally unreliable because of cross-contamination? The possible effects of cross-contamination on the Pu data are discussed in detail in Appendixes A and B and it was concluded that cross-contamination would not significantly affect most of the key samples. Additional samples were collected from soils, stream sediments, and wells by EPA scientists and particular care was taken to avoid cross-contamination. Pu concentrations in these EPA samples [9] were in concentrations similar to those found in the "6-month" study samples--thus tending to support the conclusions based on other considerations that cross-contamination of the samples did not significantly affect the Pu data from the KDHR's "6-month" study. Samples collected from the wells by the site operator also contained Pu both in the suspended solids and in the soluble portion, further supporting the quality of the KDHR data.

In brief, the samples were collected by experienced health physicists who were familiar with environmental sampling methods, the site, and operations there. More importantly, the Pu data tell a coherent story and independent Pu data collected by other scientists support this story. Therefore, it is believed that the 50 Pu analyses from the "6-month" study represent an excellent set of reconnaissance samples which cover most of the pathways for the movement of transuranium nuclides in the environment at Maxey Flats.

5.3 Other Radiochemical Data

The KDHR collected samples of liquids from approximately 40 storage tanks which contained leachates pumped from the trenches as part of the site water management program. Therefore, it is believed that these analyses represent the radiochemical character of the leachates in the trenches. Specific radionuclides detected by the gamma scan

included Mn-54, Co-60, Zn-65, Ru-106, Sb-125, Cs-134*, and Cs-137. technique included Mn-54, Co-60, Zn-65, Ru-106, Sb-125, Cs-134*, and Cs-137. They were also analyzed for Pu-238 and Pu-239, the results of which are presented in Table II. The analyses were performed on whole samples evaporated to dryness by USEPA's Eastern Environmental Radiation Facility (EERF), Montgomery, Alabama, using the analytical techniques described by Strong and Lieberman [27].

More than 300 different radionuclides reportedly have been buried at Maxey Flats [22]. Many of these have short half lives or are in small quantity. However, the following radionuclides which could significantly affect the hazard potential of any discharges from the site have been identified in the leachate or in monitoring and special study samples: H-3, Mn-54, Co-58, Co-60, Zn-65, Sr-89, Sr-90, Ru-106, Sb-125, I-131, Cs-134, Cs-137, Ac-228, U-234, U-238, Pu-238, Pu-239. These latter radionuclides have played an important role in (1) serving as a double tracer to confirm transuranium contamination** at some sample stations and (2) interpreting hydrogeological phenomena and potential pathways.

6. INTERPRETATION

6.1. General

Sixty samples were collected for Pu analysis during the KDHR's "6-month" study. They were not, however, random samples. The sample plan was designed specifically to determine if any of the transuranium nuclides were migrating--and if so, along what pathways, how much, and how far. The actual sampling points which were selected were at or near places where: (1) significant radioactive contamination had been found earlier; (2) significant alpha contamination had been found earlier; or (3) surface or subsurface water flowed from the site.

The E-series wells were specifically sampled because it was considered unlikely that contamination would have migrated to them. Therefore, detecting Pu in them would be considered highly significant.

The presence of Cs-134 in samples indicates contamination from reactor wastes.

** Unless specifically stated otherwise, "contamination" and "transuranium contamination" refer to Pu (or other radionuclide) contamination from a source other than fallout and specifically the wastes at the site.

TABLE II

CONCENTRATION OF ^{238}PU AND ^{239}PU AND RATIO OF ^{238}PU TO ^{239}PU OF LEACHATES AND LEACHATE-CONTAMINATED LIQUIDS IN TANK STORAGE AT THE MAYEY FLATS BURIAL FACILITY

	^{238}Pu (DPM/ML)	^{239}Pu (DPM/ML)	$^{238/239}$Pu (RATIO)
HIGHEST VALUE	3293	163	97.3
LOWEST VALUE	.42	NOT DETECTED	1
AVERAGE VALUE (UNWEIGHTED BY VOLUME)	214	9.02	23
AVERAGE VALUE (WEIGHTED BY VOLUME)(1)	217	10.12	22.6

(1) THE DATA REPRESENT INDIVIDUAL PU ANALYSES FROM 39 TANKS CONTAINING APPROXIMATELY 2.5 MILLION LITERS OF LEACHATE WITH WIDELY VARYING CONCENTRATION. THEREFORE, AN AVERAGE VALUE WEIGHTED BY VOLUME OF LIQUID WAS ALSO CALCULATED.

In interpreting the occurrence of Pu at Maxey Flats, it is necessary to discriminate between several types of contamination. There are: (1) contamination from atmospheric fallout and (2) contamination from a source other than fallout (i.e., the wastes and within the latter case; (2a) contamination resulting from surface spills and waste management operations; or (2b) contamination resulting from natural migration from the trenches. The data and methods used in making these discriminations were the Pu-238 and Pu-239 concentrations, the Pu-238/239 ratio and the type and location of the samples. These tools were used successfully to discriminate between (1) fallout and (2) contamination from a source. For those samples which were determined to be contaminated by ratio or concentration of Pu-238 and Pu-239, the location of the sample and its position within or relation to the hydrogeologic system were used so far as possible to discriminate between (2a) contamination from spills and operations and (2b) that from migration or movement via the water pathways.

6.2. Background and Contamination Levels

Background levels from atmospheric fallout of Pu (including Pu originating from weapons testing and SNAP generator burn-up) in the Kentucky-Ohio-Indiana area of the central United States are commonly around .02 dpm/g for Pu-239 in surface soils*; Pu-238 concentrations are commonly 4-8% of Pu-239 levels; with a resultant Pu-238/239 ratio of .15 or less [28].

To establish background levels for the "6-month" study, the KDHR collected and analyzed ten soil samples for Pu-238 and Pu-239. Six of these samples were collected in Frankfort, Kentucky, (approximately 130 air km from Maxey Flats) and analyzed by the LFE Laboratory. Four samples were collected from an unused trench at Maxey Flats and were analyzed by HSL. The background Pu data are presented in Table III. The six samples from Frankfort contained low concentrations of Pu-238 and Pu-239 with Pu-238/239 ratios which are in general agreement with regional fallout data and would not indicate contamination.

However, the four samples from the unused trenches contained unusually high Pu-238/239 ratios (mean ratio was 1.4). A recheck of the laboratory analysis worksheets and the U-234/238 ratios (approximately 1) indicate that the analyses were normal in all

* The reference gives a range of .02 - .05 dpm/g for background Pu-239 concentration. As can be noted in Table III, the mean Pu-239 concentration in the Frankfort background sampes was less than .001 and in the Maxey Flats samples, which show signs of contamination, less than .03.

TABLE III. CONCENTRATION OF ^{238}PU AND ^{239}PU AND RATIO OF ^{238}PU TO ^{239}PU IN CONTROL OR BACKGROUND SAMPLES COLLECTED AT FRANKFORT, KENTUCKY, AND AT MAXEY FLATS BY THE KDHR DURING THEIR "6-MONTH" STUDY WITH A BRIEF STATISTICAL SUMMARY.

SAMPLE/S NUMBER	^{238}Pu (dpm/g)	^{239}Pu (dpm/g)	^{238}Pu/^{239}Pu (Ratio)
F-1	.0017 ± 68%	.00844 ± 28%	.1
F-2	.00098 ± 88%	.0122 ± 20%	.1
F-3	.00096 ± 75%	.00594 ± 25%	.2
F-4	.000	.0116 ± 25%	<.1
F-5	.00000	.00716 ± 42%	<.001
F-6	.0018 ± 50%	.00792 ± 42%	.2
FF BKG	.040 ± 10	.015 ± 20%	3.7
KY BKG	.040 ± 11%	.025 ± 16%	1.2
KY BKG	.024 ± 17%	.040 ± 14%	.6
KY BKG	.028 ± 7%	.016 ± 6%	.8
STATISTICAL PARAMETERS			
FRANKFORT AREA SAMPLES (F-1 thru F-6) (1)			
NUMBER OF SAMPLES	6	6	6
RANGE	.00000 to .0018	.00594 to .0122	<.001 to .2
MEAN	.00089	.0085	.15
AVERAGE	.00097	.00754	.15
MAXEY FLATS SAMPLES (KY BKG) (2)			
NUMBER OF SAMPLES	4	4	4
RANGE	.024 to .040	.015 to .036	.8 to 2.7
MEAN	.030	.027	1.4
AVERAGE	.026	.028	.99

(1) ANALYSES PERFORMED BY LLL LABORATORY, RICHMOND, CALIF.
(2) ANALYSES PERFORMED BY ISL, IDAHO FALLS.

respects [29]. The possibility of sample contamination during collection and preparation prior to shipment was considered and is possible. An alternate possibility is that environmental concentrations of Pu are slightly but perceptibly elevated in and around the burial facility and that the general geologic framework is contaminated as a result of burial operations. This latter case would explain the contamination found in the background samples collected at Maxey Flats, in the sample from an unused trench (No. 151) and the sample across No Name Hollow from the site (No. 118). If the Pu-238 data for the entire suite of 50 samples is plotted on probability paper in the same order as on Figure 6a, a group or "family" of possibly "lightly contaminated " samples which lies between No. 128 and No. 156 is quite noticable. The fact that the background samples collected at Frankfort well away from the burial site contained more or less regional fallout background levels of Pu may also support the theory of a generally higher level of Pu at Maxey Flats.

It was concluded that the four "background" samples from Maxey Flats were collected from contaminated soil and do not represent fallout background levels of Pu.

To avoid, so far as possible, controversy as to whether a sample was contaminated by wastes or fallout, concentrations and a ratio for Pu-238 and Pu-239 were chosen which, it is believed, are high enough so that few will question that they represent contamination from the wastes. A sample was interpreted as being contaminated by wastes if: (1) the Pu-238 concentration was 0.1 d pm/g (50 times greater than background); (2) the Pu-239 concentration was 0.1 dpm/g (5 times greater than background; and (3) the Pu-238/239 ratio was 0.9 (6 times greater than background). *

Inspection of the data in graphical form also helped to establish the boundary for contamination. Histograms of the Pu-238 and Pu-239 concentrations and Pu-238/239 ratios are presented in Figures 6a, 6b, and 6c. For Pu-238 (Figure 6a), a break between fallout and contamination can be made between Samples 130 and 148 which, it is felt, is quite conservative. For Pu-239 (Figure 6b), no distinctive break is apparent. However, to choose a conservative level, the break is made at .1 dpm/g between Samples 125 and 149. For the Pu-238/239 ratio (Figure 6c), a break occurs between Samples 119 and 118 which appears to satisfy the need for both a conservative and natural break in the data to pick the fallout-contamination boundary.

* The Pu-238/239 ratio of 0.9 is not the calculated ratio between Pu-238 and Pu-239 in (1) and (2) just preceding but was chosen by the author for use in this paper and for reasons described in the following paragraph. Regardless, it is a very defendable ratio for indicating contamination from a source other than fallout.

After criteria were established as to what concentrations and ratio of Pu-238 and Pu-239 reasonably indicated contamination from wastes, an evaluation of the degree of Pu contamination in and around Maxey Flats was possible. Inspection of the basic data in Table I and Figures 6a, 6b, and 6c shows that of the 50 samples (1) 43 are contaminated based on their Pu-238 content; (2) 28 are contaminated based on their Pu-239 content; and (3) 46 are contaminated based on their Pu-238/239 ratio.

Intercomparison of Pu-238, Pu-239, and Pu-238/239 data within each sample (Table I) strongly reinforces the fact that virtually all (49 out of 50) of the samples collected are contaminated with Pu from a source other than fallout. It is also apparent that there is much more Pu-238 in the Maxey Flats soils than is usual and that the ratio of Pu-238/239 in these soils is unusually high for environmental samples in this area of the United States. Shipping records show that several major shippers of Pu-contaminated waste generate wastes which are rich in Pu-238 and which have high Pu-238/239 ratios [6].

6.3. The Leachates, a Mobile Source

In the trench dewatering portion of the water management program, leachates have been pumped from the trenches into steel storage tanks and then evaporated. Ten million or more liters of leachates have been pumped from the trenches to date. More than 2.4 million liters of leachates were stored in the tanks at the time of the "6-month" study. Total samples of the liquid, including suspended and dissolved fractions, were collected from 44 of these tanks and analyzed for Pu by EPA's Eastern Environmental Research Facility; these data are summarized Table II.

Although the liquids in tank storage are not representative of the leachates under in situ conditions, they can furnish useful information about the availability of Pu for migration. From a summary of these data (Table II.) it can be seen that the leachates in the trenches may contain, on the average, 217 dpm/ml Pu-238 and 10.12 dpm/ml Pu-239 with a Pu-238/239 ratio of 22.6. There is, without question, Pu-238 and Pu-239 in the leachate available for transport, regardless of whether it is suspended or dissolved and there are indications that some of the Pu may be in solution based on samples collected from the test wells (see discussions in Section 6.6 and 6.7). The very high ratio of Pu-238 to Pu-239 is noteworthy.

Each trench and the waste buried therein has a unique inventory of Pu in a unique waste matrix. It may, therefore, be a unique source term which will produce/ a unique leachate in the trench/. However, as these leachates migrate downward into the deeper subsurface formations, they will be subject to mixing, dispersion and dilution

occurring within the various perched and regional waterbearing formations. The end result may well be the discharge from the subsurface of a contaminated ground-water which reflects the average composition of all the leachates but possibly with several subgroups.

6.4. Other Sources of Contamination

There are potential modes of contamination at Maxey Flats other than the hydrogeologic pathways described in the following subsections. The Pu-contamination seen in the surface and near surface samples at Maxey Flats could have originated from: (1) surface spillage during general operations; (2) surface spillage during trench dewatering operations; (3) leachate which overflowed from the trenches and then infiltrated downward; (4) the use of previously contaminated fill to cap the trenches; or (5) cross-contamination by using contaminated sampling equipment. It is acknowledged here that some of the contamination in some of the samples possibly could have been caused by one or more of the five modes. Therefore, the author evaluated the potential for the various modes of contamination to affect the major points of this paper and to alter its major conclusions. The evaluation of cross-contamination by a dirty sampling device is included in Appendixes A and B.

If one takes a piece of Pu data, waves the spectre of contamination in the air, and considers no other points than, "what if," one can say contamination may possibly affect the data. If, however, one considers the radiochemical data, hydrogeological data, operations, the configuration of the sampling devices, and a number of other factors all together as they affect the quality of the Pu data, it is possible to come to the reasoned conclusion that the sources of contamination do not significantly affect the Pu data and do not rule out or lessen the importance of interflow and subsurface migration as potential pathways for Pu migration.

The lack of vertical stratification of Pu concentrations in the cores appears to minimize causes 1, 2, and 3 (surficial origin). A search of the literature has failed to identify any pertinent recorded instances where significant amounts of Pu have migrated downward from land surface greater than 30 cm by intergranular movement.* If the Pu in the leachate were sufficiently mobile to migrate downward 90 cm without significant vertical attenuation (causes 2 and 3; surficial origin), it should also be capable of migrating laterally without attenuation--thus

* The migration of Pu in the Z-9 trench at ERDA's Hanford facility [30] did not seem comparable to the situation at Maxey Flats. Downward migration of a few meters along fractures was reported in the welded tuffs at Los Alamos, N. M. [31].

reinforcing the possibility of Pu migration via the interflow pathway. Recent data from the E-series wells in which Pu was found to be in solution [7] strongly supports the conclusion that the Pu is indeed in a sufficiently mobile form to migrate via interflow through the soil and through the subsurface.

It has been suggested that the surface contamination is a result of using soil and fill which was contaminated by spillage during general burial operations and possibly from surface spillage during trench dewatering operations [6, 7]. Acceptance of this theory (cause 4) would require identification of a large source of contaminated soil and fill. From this, the questions follow where did this Pu-contaminated soil/fill come from and how was it contaminated during routine burial operations?

It has been suggested that the Pu contamination detected in the test wells was the result of contamination running into the well from land surface and or by cross-contamination during sampling. McCullough reported that the wells were constructed to prevent downward leakage [32]. Even so, the possibility that downward leakage may have occurred along a void between the grout and the wall of the wellbore has been suggested. However, the probabilities seem low that the grout would be bad on all nine wells (especially well 3E which was grouted from 0 to 18 m). It seems less probable that sufficient Pu contamination was spilled around the well for it to migrate 15 to 27 m undiluted and unattenuated. Further, four of the test wells (1E, 2E, 6E, and 8E) are located away from the operational areas where spillage should not occur. Cross-contamination is discussed in detail in Appendixes A and B and Section 6.7.

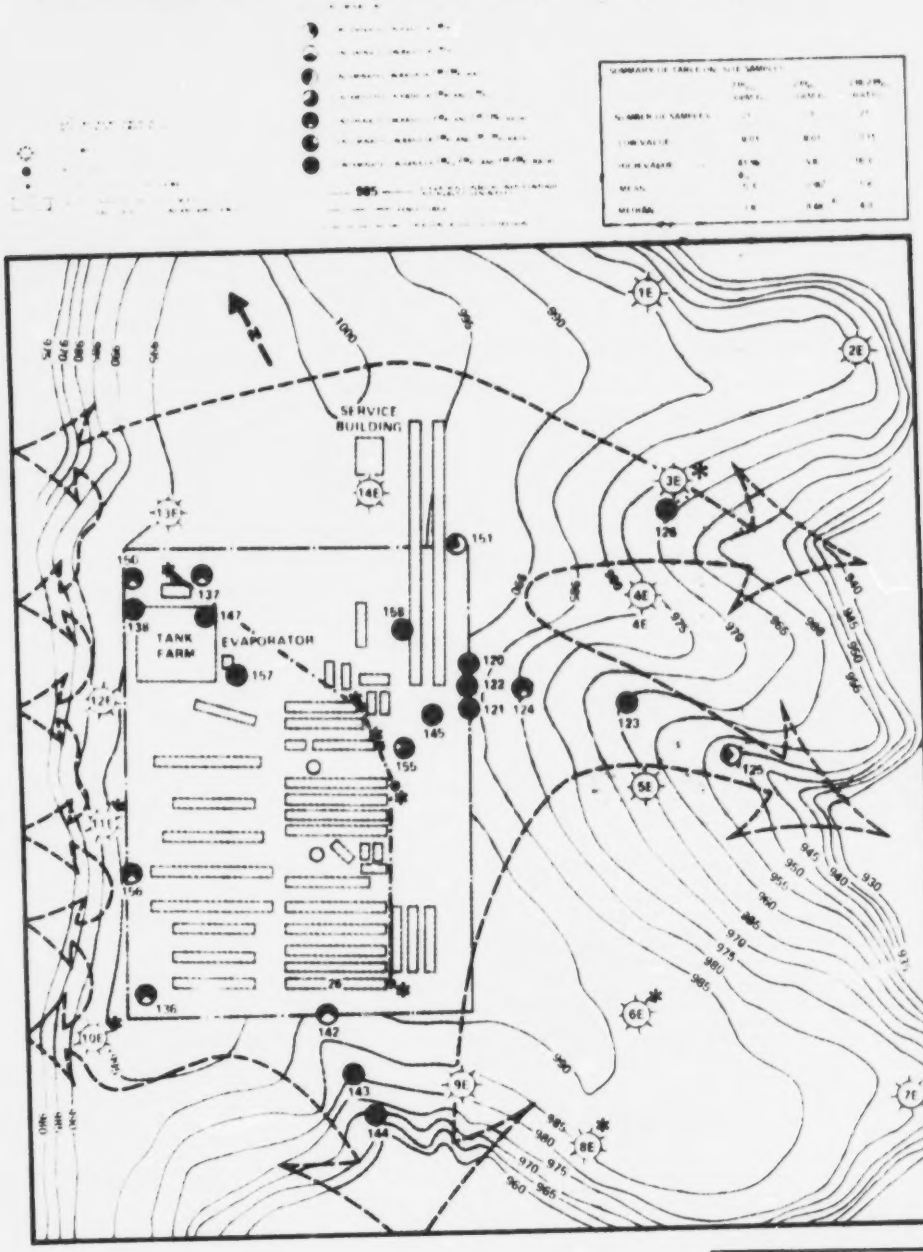
To recapitulate, it is recognized that Pu contamination from operations and sampling may have occurred; but it would not significantly affect the conclusions that (1) there is widespread Pu contamination in and around the site and (2) it is also very possible that Pu may be migrating via the water pathways.

6.5. Surface Runoff Pathway

To determine whether or not Pu contamination was moving via surface pathways at Maxey Flats, surface soil samples were collected in the burial/operational area, along major drainageways from the site, and at other selected points. Figure 7 shows the location and Pu values of surface and near-surface soil samples collected close-in to the burial

The migration of Pu in the Z-9 trench at ERDA's Hanford facility [30] did not seem comparable to the situation at Maxey Flats. Downward migration of a few meters along fractures was reported in the welded tuffs at Los Alamos, N. M. [31].

FIGURE 7. LOCATION AND SUMMARY OF CONCENTRATION OF ^{238}PU AND ^{239}PU AND RATIO OF ^{238}PU TO ^{239}PU IN "ON – SITE" SOIL SAMPLES COLLECTED AT MAXEY FLATS BURIAL FACILITY BY KDHR IN THEIR "6 – MONTH" STUDY. LOCATIONS OF SOIL CORE SAMPLES DISCUSSED UNDER "INTERFLOW" AND SUSPENDED SEDIMENT FROM TEST WELLS DISCUSSED UNDER "SUBSURFACE" ARE ALSO SHOWN. (ADAPTED FROM MAPS BY EMCON, 1973, AND CLARK, 1973.)



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area. Figure 8 is a smaller scale map showing the location and Pu values of soil and sediment samples collected "offsite", well outside of the controlled area. Inspection of the Pu values in these two figures shows that all of the "on-site" samples and 8 out of 9 "off-site" samples were contaminated.

Twenty-one soil samples were collected within or close to the controlled (fenced) operational area and were analyzed for Pu-238 and Pu-239. Analyses of 15 soil cores and 19 well samples collected within the same area are also available but are discussed separately.

A summary of the "on-site" Pu data is presented in Figure 7. Based on their Pu-238 and Pu-239 contents and their Pu-238/239 ratios, all samples but possibly one are contaminated. This latter sample, No. 151, was collected from an unused trench. It may be contaminated if judged on Pu-238/239 ratio alone. However, the Pu-238 and Pu-239 values are low enough so that the error of analysis might affect the ratio. There can be little doubt, however, that there is widespread "on-site" surface contamination.

The spatial distribution of the "on-site" sample locations is shown in Figure 7. Areas where surface runoff is likely to occur are also shown schematically. The "on-site" surface samples are so distributed that it must be concluded that Pu is moving via the surface pathway. The original source of the surface contamination cannot, however, be determined. Potential sources include leachate overflowing from the trenches, evaporator fallout, spillage during routine operations, spillage during dewatering operations, lateral migration through the soil to the surface, or a blend of all of these sources.

The location of the soil cores and the monitoring wells which are discussed later are also shown in Figure 7.

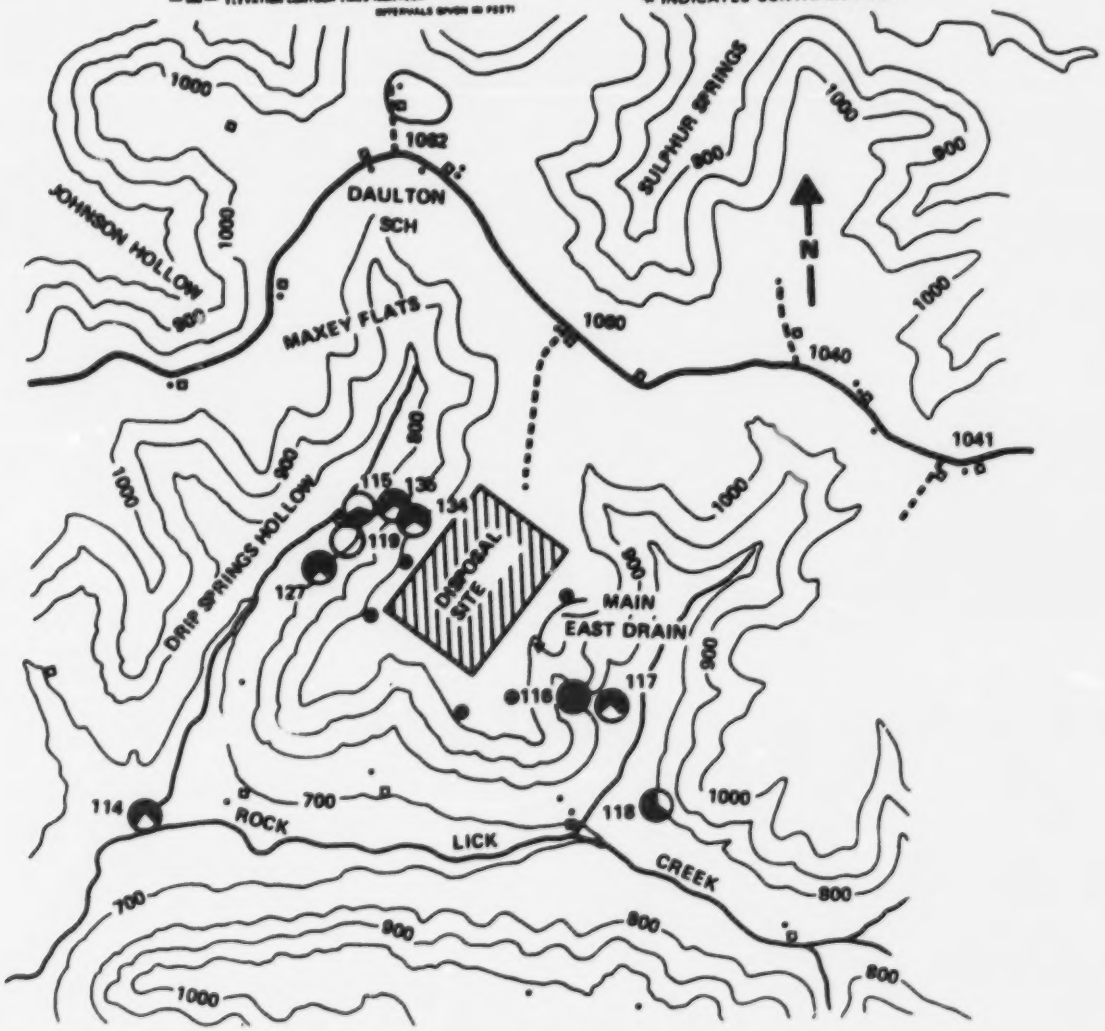
The locations of the off-site samples are shown in Figure 8. A brief discussion of the Pu values of each "off-site" sample point and their relationship to the hydrogeology and potential pathways at Maxey Flats follows:

Sample 114: Contaminated stream sediment which could represent contamination from any or all of four pathways (surface, interflow, subsurface, and evaporator) discharging into Drip Springs Hollow. Located more than 1500 m from the burial trenches via the shortest surface flowpath.

FIGURE 8. LOCATION, CONCENTRATION OF ^{238}PU AND ^{239}PU, AND RATIO OF ^{238}PU TO ^{239}PU IN "OFF- SITE" SOIL AND SEDIMENT SAMPLES COLLECTED AROUND MAXEY FLATS BURIAL FACILITY BY KDHR IN THEIR "6-MONTH" STUDY. (ADAPTED FROM U.S. GEOLOGICAL SURVEY FARMERS, KY., AND PLUMMERS LANDING QUADRANGLE TOPOGRAPHIC MAPS.)

SAMPLE POINT	^{238}Pu (DPM/G)	^{239}Pu (DPM/G)	^{238}/^{239}Pu (RATIO)
114	*0.57	0.073	*7.81
115	0.076	*0.298	0.254
116	*1.92	*0.99	*1.93
117	*0.18	0.044	*4.09
118	*0.13	*0.14	*0.96
119	0.034	0.089	0.389
127	*0.50	0.05	*10.00
134	*0.15	0.01	*15.00
135	*0.11	0.01	*11.00

* INDICATES CONTAMINATION



Samples 115 and 127: Contaminated pond sediment and soil, respectively, which could represent contamination from any or all of the previously discussed pathways down the west drainage way from the site. Contamination from surface runoff is strongly suspected for both. At No. 115, the pond may also be spring-fed, the source of which could either be interflow, subsurface flow, from the bedrock or alluvium, or a blend of both. No. 127 may also be fed by interflow, subsurface flow, or a blend of both.

Samples 116 and 117: Contaminated stream sediment which could represent contamination from all pathways down the main east drainage way from the site. Contamination from surface runoff is almost certain at both points. However, No. 116 is a sediment collected below a spring which may be fed by interflow, subsurface flow, or a or a blend of both.

Sample 118: Decomposed vegetation from a "sulfur" spring (Ohio Shale spring?) across No Name Hollow from burial site; it is separated from the site hydrogeologic system. The atmospheric pathway should be the only pathway which could contribute Pu. From its location, the sample should represent background or close to it. It is contaminated by the criteria used herein and fallout from the evaporator plume is a possible source of the contamination.

Sample 119: Sediment from pond in Drip Spring Hollow beneath burial site. It may be fed by surface runoff, interflow, subsurface flow from the bedrock and alluvium, or a blend of all these.

Samples 134 and 135: Slightly contaminated soil. Samples 134 and 135 are particularly interesting because they are adjacent to Well 12E, in which a zone of radioactive contamination was detected at a depth of approximately 13-14 m by geophysical logging. Subsequent bore hole geophysical logging of the hole by the U.S. Geological Survey using the gamma spectral method identified the presence of specific waste-associated radionuclides Co-60, Cs-134, and Cs-137 [33]. Three factors suggest that the contamination is from subsurface flow: (1) the description of the sample locations, "back into the hill under a shale outcrop;" (2) the sample locations are down the hill at the same approximate elevation as the zones of contamination which were detected in Test Well No. 12 [17], would be expected to outcrop; and (3) the Pu-238/239 ratios are relatively high, as were the ratios in other surface samples from the test wells. Contamination of these samples by surface runoff and interflow is possible. No. 134 was not on a surface runoff path, however, but could be on an interflow path.

6.6. Interflow Pathway

A well-developed soil zone, 1-5 m thick, covers Maxey Flats and its slopes and is present in the trench area of the site. In addition, the trenches are capped with mixed soil and shale which was excavated from them. This cap material is completely disturbed and disaggregated and has many of the properties of soil. Because some of the trenches filled to ground-level or near-ground-level with leachate, interflow, or the lateral migration of water through the soil, is a potential pathway for the migration of Pu. Leachates overflowing from the trenches or discharging to land surface or soils on the slopes below the site could also contribute contamination to the interflow pathway.

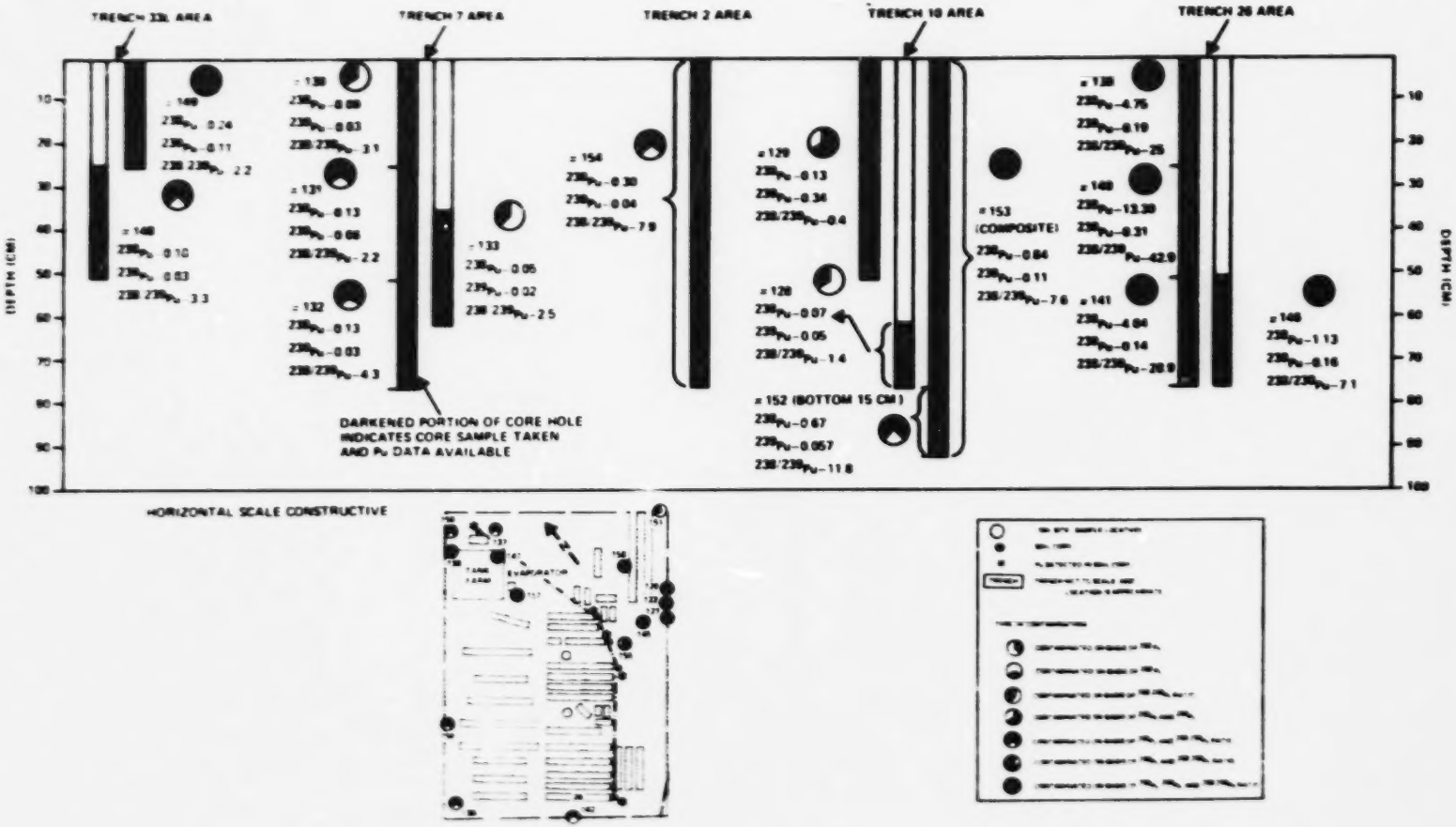
It is difficult to determine whether the movement of Pu by interflow is occurring along the flanks of the site because of the intimate intermix of the surface, interflow, and subsurface pathways in this area (see inset, Figure 3). However, on top in the relatively flat or gently sloping trench area, the pathways are less mixed and a preliminary evaluation of migration by interflow seems possible.

Fifteen cores were taken from soil or cap material close to the trenches and range in depth from land surface to 90 cm (see Table I for a description of their location). The spatial relationships of these cores and their Pu concentrations are shown in cross-section in Figure 9. Observations which can be made about the cores, their Pu content, and their hydrogeologic relationships follow:

- Most of the cores are believed to be trench cap material based on their location and depth.
- All 14 of the cores are contaminated based on the general criteria stated in Section 6.2.
- There was no apparent diminution or "plating out" of Pu with depth, although experience elsewhere shows that little or no Pu migrates downward below 15 cm (see discussion in Section 7.1).
- The Pu-238/239 ratios for cores near Trench 26 are very high and resemble some of the ratios in the leachate--possibly suggesting a common source.

We know from operational reports and a special study [21] that some of the trenches were filled, or nearly so, with a potentially mobile source of Pu contamination--the leachate. The cores were collected

FIGURE 9. SCHEMATIC CROSS- SECTION OF TRENCH AREA OF MAXEY FLATS BURIAL FACILITY SHOWING THE CONCENTRATION OF ^{238}PU AND ^{239}PU AND RATIO OF ^{238}PU AND TO ^{239}PU AND THE SPATIAL RELATIONSHIPS OF THE SOIL CORES COLLECTED IN THE KDHR 6-MONTH STUDY.


TRENCH 33L AREA
TRENCH 7 AREA
TRENCH 2 AREA
TRENCH 10 AREA
TRENCH 26 AREA
DEPTH (CM)
DARKENED PORTION OF CORE HOLE INDICATES CORE SAMPLE TAKEN AND Pu DATA AVAILABLE
HORIZONTAL SCALE CONSTRUCTIVE

at the east or southeast end of the trenches which one can logically infer to be downgradient from the trenches assuming that the dip of the beds and mounding of leachate in the trenches influenced the direction of water movement in the trench area. This would be the direction then in which these leachates would migrate, if they migrated. The following hydrogeologic information suggests that Pu could migrate from the trenches via the interflow pathway: the trenches were filled to land surface; the soils had some degree of permeability; and a hydraulic gradient existed--therefore interflow or lateral migration of the leachate through the soil should have taken place.

Based on the core data, there is Pu contamination of the soil and cap material in the trench area at Maxey Flats down to a depth of 75-90 cm. This contamination appears to be relatively uniform from land surface downward to at least 90 cm depth with little sign of stratification or preferential distribution. Further, it can logically be inferred that the contamination extends below 90 cm and possibly to the base of the weathered zone. If there is no obvious stratification of Pu down to 90 cm, there is no good reason to limit or fix a point below 90 cm where stratification should begin except at some natural hydrogeologic boundary. The base of the fill or weathered zone is such a natural boundary. To use a specific example, in the Trench 26 area, the probability that the concentration of Pu drops to "zero" at 92 or 95 cm is low when it is obviously contaminated to the 1-4 dpm/g level at 90 cm.

The core data together with the hydrogeological information available suggest a situation where the trenches filled with leachate and the leachate migrated laterally through the soil and cap material away from the trenches or overflowing from the trenches had saturated the areas sampled. One could reasonably expect to find stratification of Pu concentrations downgradient away from the trenches.

While lateral migration was occurring, downward infiltration could be expected through the bottom of the trenches into the Nancy and Farmers Members because the trenches extend below the weathered zone into the jointed Nancy Member.

Based on information about wastes buried at other sites it is believed that decontamination solutions are buried with low-level wastes. If confirmed, these decontamination solutions possibly acting in conjunction with organic acids which are commonly generated in land fills such as Maxey Flats could act to put Pu and other radionuclides into a complex or anionic form which could readily migrate with the movement of the leachate and soil water. Although the same information is not available for the surface soil and soil core samples, the presence of Pu in soluble form in the wells [7] supports the hypothesis that it may be present in soluble form in the near surface soil zones, also.

It has been suggested that the Pu contamination seen in the surface and near surface samples at Maxey Flats could have originated from other sources (Section 6.4). This possibility is recognized; it does not, however, lessen the possibility that the migration by interflow may have occurred.

6.7. Subsurface Pathway

In 1973, EMCON Associates, Inc., a geological consulting firm, constructed 14 monitoring wells around the perimenter of the controlled area of the burial site. These wells were constructed and completed at different depth intervals for the primary purpose of monitoring for contamination in the different subsurface formations beneath the site. They also had the secondary purpose of identifying water-bearing horizons or aquifers beneath the site. Excellent information is available on the character and depth of the different subsurface formations because core samples of the formations penetrated by the wells are available [18]. Figure 10 and Table IV give details on Wells 3E, 6E, 8E, 10E, and 11E.

The following points argue against the introduction of significant Pu-contamination having been introduced into the wells by means other than subsurface migration:

- The Pu-238/239 ratios found in some of the wells are higher than ratios generally found at land surface.

- The cross-contamination mechanism does not seem adequate to produce the concentrations and ratios of Pu actually observed in the wells (see Appendixes A and B).

- At least some of the wells in which Pu contamination was detected were not on paths where overland runoff from contamination areas could easily flow around the wells and run down the annulus of the well to cause contamination.

- Independent sampling by three organizations (KDHR, EPA's RNEB, and the site operator) found Pu contamination in nine wells (1E, 2E, 3E, 6E, 8E, 10E, 11E, 12E, and 13E) in 20 out of 20 suspended sediment samples reported on thus weakening the arguement for cross-contamination (Table V).

- Other radioactive contaminants were found in the wells (H-3, Co-60, Cs-134, Cs-137).

- Pu has been found in solution in four samples from two wells.

FIGURE 10. SCHEMATIC OF CONSTRUCTION OF TEST WELLS AT MAXEY FLATS FOR WHICH PU DATA ARE PRESENTED IN THIS REPORT. (BASED ON INFORMATION PRESENTED BY EMCON)

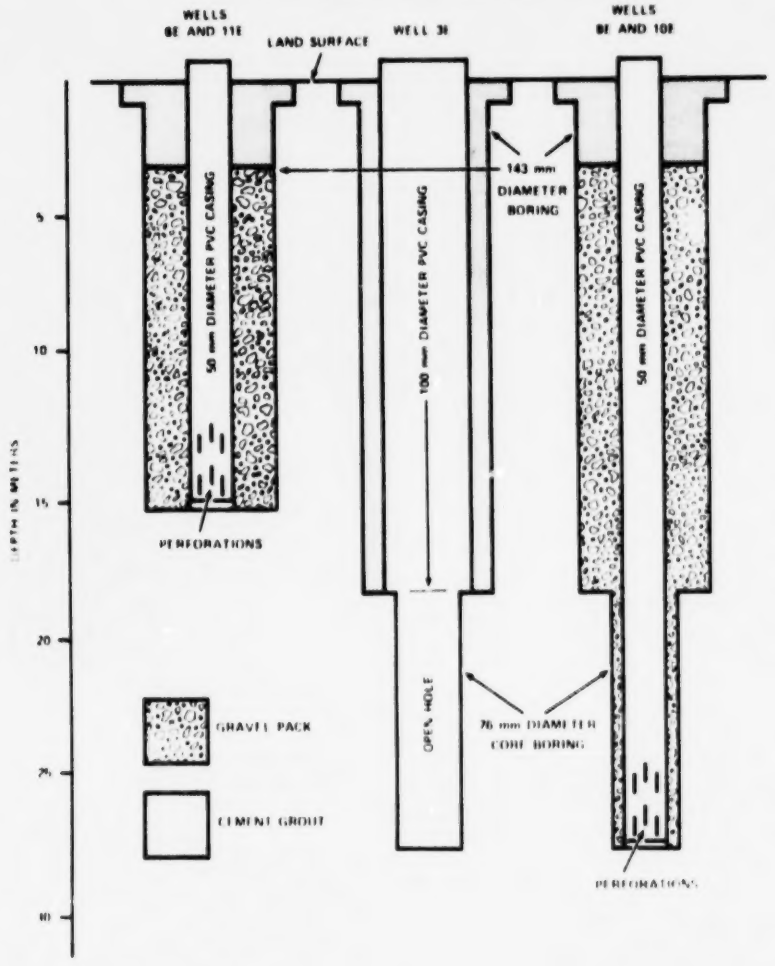


TABLE IX. SUMMARY OF WELL CONSTRUCTION, GEOLOGIC, AND HYDROLOGIC DATA FOR TEST WELLS AT THE MAXEY FLATS BURIAL FACILITY WHICH WERE SAMPLED FOR PU BY KDHR DURING THEIR "6-MONTH" STUDY AND BY RNEB-CINC.

				BASIN UNITS			BASIN UNITS (WATER-BEARING)				
WELL NUMBER	TOTAL DEPTH (FT)	TOTAL DEPTH (M)	ALTITUDE OF WELL (FT MSL)	ALTITUDE OF WATER (FT MSL)	ELEVATION OF WATER (M MSL)	DEPTH (M)	DRAINABLE FORMATIONS	RELATIVE WATER PRODUCTIVITY (2) OF WELL	DRAWDOWN INTERVAL (M)	FORMATIONS PENETRATED BY WELL
6	77.4	23.6	911.4	25.6	265.4			MODERATE	0.15	SAXEY, FARMERS (UPPER)
8A	43.2	13.2	926.7	33.1	313.6			MODERATE	0.3	SAXEY, FARMERS (UPPER)
9A	77.4	23.6	926.9	24.2	298.1	22.9-23.8	FARMERS	LOW	0.3	SAXEY, FARMERS, HENLEY, SUNBURY (UPPER)
10A	77.4	23.6	920.1	27.4	302.3	17.1-17.9 21.0-21.6 25.6-25.9	FARMERS FARMERS SUNBURY	LOW	0.3	SAXEY, FARMERS, HENLEY, SUNBURY (UPPER)
11A	43.2	13.2	921.3	15.4-15.5	305.7-305.9			MODERATE	0.3	SAXEY, FARMERS (UPPER)

(1) Based on loss of drilling fluids during coring and drilling operations.

(2) All of the formations penetrated are aquitards, the test wells do not produce water like a water well does. The relative productivity of the test wells is defined as follows: low drawdown recovery less than 30 cm after 2 weeks, moderate drawdown recovery approximately 30 cm in 2 weeks, and good, drawdown recovery 180 cm in 5 days.

Table V. Summary of concentration of Pu-238 and Pu-239 and ratio of Pu-238 to Pu-239 in suspended solids and soluble portions of water samples collected from test wells at Maxey Flats burial facilities. (2)

	Suspended Solids	Soluble
Number of analyses	20	15
Number of Samples Contaminated with Pu (On Any Basis)	20	4
Number of Samples Contaminated with Pu (On Basis of Pu-238)	20	2
Number of Samples Contaminated with Pu (On Basis of pu-239)	11	2
Number of Samples Contaminated with Pu (On Basis of Pu-238/239 Ratio)	20	NA (1)
Number of Wells Sampled	9	7
Number of Wells contaminated with Pu (On Any Basis)	9	3

	Suspended Solids (3)			Soluble (4)		
	Pu-238 (dpm/g)	Pu-239 (dpm/g)	Pu-238/239 (Ratio)	Pu-238 (dpm/l)	Pu-239 (dpm/l)	Pu-239/239 (Ratio)
High Value	33.3	1.2	71.7	28.9	?.3	13
Low Value	0.18	0.013	0.7	0.02	0.01	NA (1)
Median Value	7.9	0.21	32.9	NA (1)	NA (1)	NA (1)
Mean Value	11.9	0.69	31.1	NA (1)	NA (1)	NA (1)

1/ Not applicable because of insufficient data.
2/ Detailed data is presented in Appendix C.
3/ Based on 20 samples.
4/ Based on 15 samples.

The Pu data from the "6-month" study and other sources when taken with the hydrogeologic data are adequate to establish that the potential for subsurface migration exists. Other radiochemical data [2, 9] also suggest that Pu may be migrating via the subsurface pathway. To help the reader understand the complex relationships between these diverse data, a 3-dimensional isometric stratigraphic panel diagram which shows the wells, the geologic formations they penetrate, the potential source term, some of the hydrology, the potential pathways, and potential zones where contaminationcan occur is presented in Figure 11. Observations made about the potential source term, the monitoring wells, and their interrelation with the subsurface hydrogeology follow:

- There were leachate-filled trenches [21] which could serve as a potential source of contamination (FACT).

- The hydrostatic head of 4-6 m of leachate [21] which existed in some of the trenches in the past could have furnished considerable driving force for this source term (FACT).

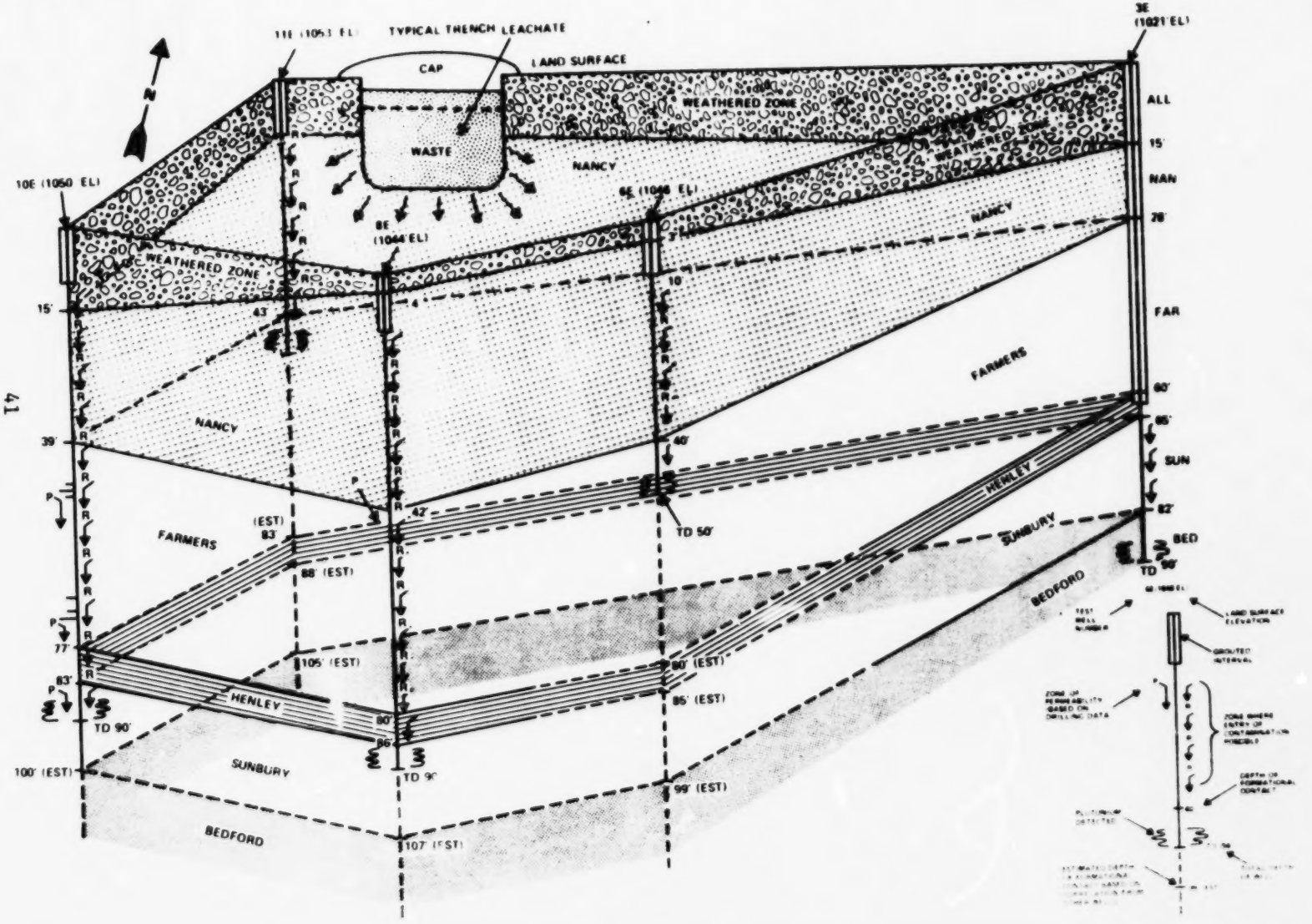
- Joints in the burial media and underlying subsurface formations [15, 14, 17, 18] are potential pathways for migration through otherwise low-permeability rocks (FACT).

- Zones of subsurface permeability have been reported [15, 18]; therefore, potential pathways do exist for downward subsurface migration of Pu (FACT).

- Water has been detected and repeatedly measured in the monitoring wells [17, 19, 34], establisheing that presence of what are believed to be perched aquifers and a regional water table. Because infiltrating precipitation is the most likely source of groundwater beneath the site--these waterbearing zones tend to support that downward movement does occur. (HYPOTHESIS held by six geologists) [15, 21, 14, 34, 17 and this report].

- Pu has been detected in suspended solids and sediments in wells 3E, 6E, 8E, 10E, and 11E in the 6-month study [2]. Pu has been detected in suspended solids and in solution in wells 1E, 2E, 3E, 6E, 8E, 10E, 11E, 12E, and 13E in other studies (FACT) [7, 9].

FIGURE 11. SCHEMATIC OF CONSTRUCTION OF TEST WELLS AT MAXEY FLATS FOR WHICH PU DATA ARE PRESENTED IN THIS REPORT.



- Based on the depth the casing was emplaced and grouted, and assuming that the grout is effective, Pu could possibly have entered the wells from the following depths and formations: Well 3E, 18-26 m from the top of the Bedford and the Sunbury; Well 6E, 3-15 m from the Nancy and Upper Farmers; well 8E, 3-28 m from the Nancy, Farmers, Henley, and Upper Sunbury (permeable zones reported at 17-18 m in the Farmers and at 25-27m in the Sunbury); Well 10E, 3-28 m from the weathered zone, Nancy, Farmers, Hanley, and Upper Sunbury; Well 11E, 3-16 m from the Nancy and the Upper Farmers (FACT) [18].

- The Pu-238 concentrations in the well samples are very high relative to all other samples collected in the "6-month" study; all fall within the 94 percentile (FACT) [2].

- The Pu-238/239 ratios in the well samples were very high relative to all other samples collected in the "6-month" study (all fall within the 94 percentile) [2]. They were also similar to ratios in the leachates, possibly suggesting a common source. Preliminary analyses from RNEB-CINC also indicate that the well samples have very high Pu-238/239 ratios (FACT) [3].

It is believed that the preceding observations strongly support the following hypothesis: (1) there is a mobile potential source of Pu-contamination (the leachates); (2) there was and possibly still is a driving force (the hydrostatic head of the leachate); (3) there are subsurface pathways for the migration of Pu (the joints); and (4) Pu was detected in the wells. Therefore, the information available suggests the possibility that Pu may be migrating from the trenches via the subsurface pathway. Additional information and studies are required to confirm this, however.

The occurrence of very high concentrations of Pu-238 and Pu-238/239 ratios in the wells compared to other samples collected at Maxey Flats (excluding the leachate samples) is significant.

6.8. Atmospheric Pathway

Volume reduction of the leachate by evaporation prior to its solidification is an integral part of the water management program for the site. The liquid to be evaporated is pumped into a settling

: The author personally made the crude but simple test for water in the wells by dropping a small pebble into the well and listening for a splash as indicating the presence of water. A splash was heard in 8 or more of the wells. Later, Zehner in his studies has routinely measured water levels with automatic water-level recorders and/or chalked steel tape.

tank, allowed to settle for several hours, pumped into the evaporator vessel, heated by a submerged propane burner, and evaporated at a rate of approximately 19 l/min. The vapor is passed through a mist eliminator and then discharged to the atmosphere through a 10 m stack. The evaporator was designed specifically for Maxey Flats operations. The evaporator "bottoms", or concentrates, are periodically pumped into a holding tank to await solidification.

Pu-contaminated liquids go into the evaporator; therefore, the vapor plume discharging from it is a potential pathway for Pu. Although possible, volatilization of Pu is not necessarily suspected. However, the discharge of extremely small particles is of concern. RNEB-CINC has detected 11 mostly long-lived radionuclides in the stack effluents including H-3, Na-22, Co-60, Sr-90, Ru-106, Sb-125, I-131, Cs-134, Cs-137, Ra-226, Ac-228, and gross alpha [5]. Pu analyses of the stack effluent are not yet available.

One sample collected during the 6-months study may be directly related to the evaporator pathway. Sample 157, which was collected from soil directly beneath a drip from the discharge stack, had 20 dpm/g Pu-228, .49 dpm/g Pu-239, and a Pu-238/239 ratio of 11.93.

The physical aspects of the evaporation process may be of concern when dealing with radionuclides which are known to occur in extremely fine particles, such as Pu-238 and Pu-239.* First, suspended solids in the liquid to the evaporated are removed by settling, a process which may not be very effective for extremely fine particles.** Any solids carried over into the evaporator vessel are subjected to considerable turbulence during evaporation by the submerged flame. The fall back of finely divided particles into the "bottoms" may not be as complete as anticipated. Second, the decontamination efficiency of the system drops off as the "bottoms" become more concentrated. Therefore, controlling the concentra-

* Volatile radionuclides are also of concern but are not the subject of this paper.

** This potential problem should be largely eliminated now because the pre-treatment of the leachate with flocculating agents is reportedly greatly reducing the suspended solids in the liquid going into the evaporator.

tion of the evaporator "bottoms" is essential to maintaining uniform decontamination efficiency in an evaporator system. In a preliminary report, RNEB-CINC noted a low correlation between the feedstock going into the evaporator and the effluent discharging from the stack [8]. Lack of precise control of the concentration of the "bottoms" is one possible answer to the apparent lack of correlation.

Contamination via the atmospheric pathway would certainly provide a plausible answer to why a number of the samples in the 6-month study appear to be "lightly contaminated" on the basis of their Pu-238 content and Pu-238/239 ratio.

7. ENVIRONMENTAL IMPLICATIONS OF MAXEY FLATS PLUTONIUM DATA

7.1. Low-Mobility Concept of Plutonium

To date, there have been few field reports that Pu has migrated when buried in the ground. To the contrary, Pu has the reputation of being sorbed on the soil within several centimeters of contact. Reichert [35] reports that by 1962, all of the radionuclides released to the seepage basins at the Savannah River Plant (SRP), except Pu had been detected in the surrounding groundwater. Fennimore [36], also at SRP, concludes that Pu does not move through soil even under a constant hydraulic head.

The studies on the migration of Pu in the environment have been influenced by its mode and place of occurrence and possibly by the type of Pu. By far, the largest source of Pu in the environment has resulted from deposition on soil and water surface due to fallout, accidents, and releases from normal operations. Therefore, much of the research in recent years has been concerned largely with the fate of plutonium deposited on soil surfaces. In addition, much emphasis has been placed upon field investigations of migratory pathways at semiarid sites. Laboratory investigations have concentrated largely on column studies of Pu leach rates and soil adsorption capabilities of a media occurring during the intergranular flow of a contaminated liquid through pulverized media.

Researchers seem to generally agree that Pu is effectively retained in the uppermost part of the soil column and is not available to infiltrating water even under humid climatic conditions.

A commonly accepted axiom is that Pu does not migrate deeper than 15 cm below the surface in soils, unless they are disturbed [37, 38, 39, 37]. Some workers [41, 42] suggest that the downward dispersal occurs by mechanical movement of Pu-contaminated particles.

Hajek [43], working on the behavior of Pu in subsurface disposal at Richland, Washington, concluded that movement by contact diffusion would be less than 10 cm after 240,000 years, and that a maximum of 0.1% of the plutonium would be leached by infiltrating ground water. Rhodes [44] reports that soil sorption of Pu was greater than 97% between pH 2 and 8. Wilson and Cline [45] were able to remove less than 1% of the Pu in a soil column using standard (agricultural) leaching techniques.

7.2 The Mobile Concept of Plutonium

A number of agents and conditions can increase the mobility of Pu when it is buried in the ground. A few of these are described briefly below.

-- Pu can, in the presence of certain agents, form complexes which are difficult to remove from solution and such complexes would not be expected to be retained by soils [46].

-- The mobility of Pu in Soils with extraction solutions has been demonstrated using an equilibrium method. It was concluded that Pu was extracted due to the formation of anionic acetate complexes* and not the effect of the extracting cations [44]. Similar results were recorded with organic solvents which are used during nuclear fuel reprocessing operations [47].

-- Pu uptake by plants may be enhanced by successive cropping and the passage of time. Although Pu uptake by perennial ryegrass was low, it increased over a two-year period. It was detectable in the third harvest of the first growing season in an acid soil and increased four-fold in the second year [48, 49].

* It is interesting to note that acetic acid is a common product of the anaerobic decomposition process in landfills.

-- The uptake of fallout Pu by clover increased 7-fold over a period of five years; the increase was believed due either to more roots coming in contact with Pu as the plants aged or that natural organic materials resulting from root tissue decay complexed with Pu [40]. (The potential complexing of Pu by plant activity may be more important than its uptake by plants).

-- Uptake of Pu was more than three times greater in a moderately acid forest soil (such as found in the humid East) than in calcareous soil (such as found in the arid West) [50].

-- Chelating agents such as DTPA (diethylenetriamine pentacetic acid) can cause a 20-fold increase in Pu uptake by alfalfa [51].

-- Studies of the effects of chelating agents on the availability and mobility of Pu in eco-systems are important in view of the commercial use of chelates in supplying micronutrients to crops and plants [42].

-- Subsurface hydrogeologic and hydrochemical parameters affect the sorptive capacity of a soil including pH, redoxpotential, Eh, and moisture content [14].

-- Soil sorption capacities are minimized if Pu transport occurs along fractures or joints rather than interstitially because of the reduction of rock or soil surface area exposed to the contaminant. Christenson and Thomas [31] have reported that Pu released to seepage basins has been able to migrate to depths of at least 8.5 m through fractures in the Los Alamos Tuff. They mentioned the possibility that organic complexes were responsible for the transport of Pu; however, they felt that the Pu movement took place along fissures.

Price [42] and Francis [41] present excellent reviews of the published studies on the distribution and fate of transuranium elements in the environment. Price concludes that the behavior and fate of transuranium elements in the environment is poorly understood. Francis concludes that the mobility of Pu after deposition on the surface of rich prairie or forested soil developed in a temperate to subhumid or humid climate is unknown.

Gera [52] more recently has introduced data from volcanic environments which suggests considerable geochemical mobility for Pu but the data are too scanty and too restricted to provide a reliable indication of

the general behavior of Pu in geologic systems. He noted that the migration of Pu colloids through geologic formations would be restricted by filtration and surface sorption, rather than by ion exchange. Gera further noted that the mechanism of infiltration in humid areas could be significantly different than in arid areas and that biologic processes could also contribute to the downward movement of Pu.

7.3. Behavor of Plutonium When Buried in a Landfill at Maxey Flats

In the previous sections, the author presented "just the facts" or what reasonably could be deduced from the facts. This section is more hypothetical in nature. Herein, an attempt is made: (1) to make the connection between a radioactive waste disposal site as presently operated in a humid climate and a common landfill garbage dump and (2) to suggest that somewhere in the processes going on within the landfill trenches at Maxey Flats and elsewhere, there is some special mechanism which is able to mobilize Pu contrary to common accepted concepts of Pu's immobility. The author would like to state specifically that he does not believe that the mechanism(s) suggested here regarding Pu's mobility in the environment apply to the disposal of Pu in a deep geologic disposal situation such as been suggested for the ultimate disposal of high-level and transuranium-contaminated wastes. Rather, the special problems addressed here are associated with the shallow disposal of Pu and other radionuclides in landfills in humid climates using present methods.

The Maxey Flats burial facility has had a number of operational problems such as trenches filling with water, erosion and subsidence of caps, and the movement and possibly migration of leachates from the trenches. These and other problems are normal to most conventional landfills in humid climates--and Maxey Flats is basically a landfill which accepts only a special type of waste. Two important causes of these problems are: (1) the landfill process which greatly alters the natural hydrogeologic system of a site and (2) the large amounts of contaminated solid waste which are placed in an environment favorable for its dissolution and dispersal.

To understand the problems at Maxey Flats and how it is possible for Pu to migrate, an understanding of what happens to waste after it is buried in a landfill is helpful. Therefore, a brief description of the "life cycle" of a landfill follows: (1) the trench is excavated, and quite often, as at Maxey Flats, from material with relatively low per-

meability; (2) the trench is filled with high porosity, permeable, compressible wastes which contain organics and a wide range of chemical forms; (3) the wastes are covered with an earthen cap which is often more permeable than the original pre-trench soil and rock, in effect, creating an infiltration gallery; (4) some of the precipitation which falls on the cap, infiltrates into the trench and soaks the wastes; (5) leaching of the waste begins, aided by the presence of organic matter, bacterial action, the formation of organic and inorganic acids, and chelating agents; (6) the trench leachate begins (i) to migrate downward and laterally because of the hydraulic head imposed by the leachate in the trench and/or (ii) to overflow at land surface in springs and seeps at some low point between the cap and the undisturbed earth; and (8) as the wastes continue to soak and leach, they compact, undermine the trench cap, increase the infiltration of water into the trench, and thereby increasing leachate generation.

Landfill leachate commonly has 50,000 - 80,000 mg/1 chemical oxygen demand, 11,000 mg/1 of volatile acids, 6,000 mg/1 of organic acids, and pH ranges of 3.7 to 8.5 [53, 54]. Constituents which possibly might release or mobilize contaminants from the waste include acetic, proprionic, isobutyric, butyric, and valeric acids. In brief, landfill leachates may contain agents capable of putting into solution generally insoluble elements such as plutonium.

Detailed analyses of the non-radioactive constituents of the Maxey Flats leachates are not available. If, however, it is recalled that the wastes entering a commercial radioactive waste burial facility may typically contain (1) 70% by volume paper and plastic material, (2) numerous laboratory animals, and (3) organic compounds, and are often packed in wooden crates and cardboard boxes (which are organic material), it can be seen that "other-than-high-level" wastes make quite a rich feedstock for generating a very potent landfill leachate. In the absence of additional information, it seems reasonable to assign at least some of the characteristics of a "typical" landfill leachate to the Maxey Flats leachates.

Preliminary analyses of leachates collected from trenches at the commercial radioactive waste disposal facility in West Valley, New York, indicate that they may be similar in character to common landfill leachates [55]. Nineteen different organic peaks were identified including acetic and butyric acids. In addition, Pu was detected in both the dissolved and suspended fractions of the West Valley leachates. In one sample, approximately 5% of the Pu in the leachate was in the dissolved fraction; the other 95% was in the suspended fraction.

As noted earlier, Pu was detected in the leachates at Maxey Flats, but the analyses were on the total leachate samples, so it is not known whether the Pu was suspended, dissolved, or both. Finding Pu in soluble form in the wells [7] makes it reasonable to suspect, however, that Pu is in dissolved form in the leachates. From what is known about the wastes buried at Maxey Flats, it seems reasonable to suspect that Maxey Flats leachates share many of the characteristics of common landfill leachates, also.

After reviewing what little is known about the behavior of Pu in the environment, it appears that the climate, the geology, and the burial conditions at Maxey Flats include many of the factors which would tend to mobilize Pu or to reduce its retention in the ground--and few factors which would tend to immobilize it. For example, it is known or it can be reasonably inferred that:

- Maxey Flats has a humid climate and acid forest soil.

- Grasses and clovers have been grown on the caps and cropped for the past several years.

- The burial media and underlying strata are jointed and fractured which reduces their effective sorptive action on contaminants passing through them.

- Pu forms complexes with volatile and organic acids, chelating agents, and decontamination solutions from nuclear facilities. The Pu complexes once formed are difficult to remove from solution and are not readily retained by soils.

- Volatile and organic acids are generated naturally in landfill leachates in large volumes. Landfill leachates commonly contain 10,000 - 15,000 mg/1 volatile acids - and it is belie ed there are similar leachates in the trenches at Maxey Flats.

- One can reasonably infer that wastes containing decontamination and cleanup solutions from nuclear facilities have been buried in the trenches.

The migration of leachates from landfills even in arid and semi-arid climates is a reported fact; in a humid climate leachate migration or movement is almost de rigueur. Referring more specifically to radioactive waste disposal sites, the migration or movement of radioactivity has been reported at the commercial burial site at West Valley, New York [56]; at the burial site at the Oak Ridge National Laboratory, Tennessee [57], and at a former AEC facility at the Palos Hills Forest Preserve (near Argonne National Laboratory), Illinois [58].

Therefore, the author believes that the widespread occurrence of Pu and its distribution in the leachates, soils, soil cores, and wells at Maxey Flats are indications that (1) Pu possibly has been mobilized by one or a combination of the above agents and (2) has migrated through the soils and subsurface formation at Maxey Flats. This report does not try to specifically define how or in what form Pu is migrating; its primary purpose is to call attention to the very likely possibly that it is migrating.

8. SUMMARY AND CONCLUSIONS

Wastes containing more then 80 kg Pu-239 have been emplaced in trenches where water infiltration has occurred. Pu and other radionuclides have been leached or released from the wastes to form a Pu-contaminated leachate. Therefore, a large source term of Pu is available at Maxey Flats in a mobile form.

There are at least four potential pathways available for the migration of Pu at Maxey Flats. These include: (1) the surface water transport of contaminants which reach land surface; (2) interflow, or lateral migration of contaminants via water movement through the soil; (3) subsurface migration of contaminants along joints and bedding planes in the underlying strata; and (4) atmospheric dispersion via the evaporator plume.

Available hydrogeologic information indicates that there are, or recently have been sufficient driving forces to cause the Pu-contaminated leachate to migrate. The hydrostatic head of 4-6 m of the leachates in some of the trenches is or was sufficient to cause lateral and downward migration of the Pu-contaminated leachates. The normal erosional processes are sufficient to cause surficial migration of Pu once it reaches land surface, either through operational activities, leakage from the trenches or lateral migration through the soil. The evaporator plume is a potential pathway for any Pu entrained therein.

Pu in above background concentrations has been detected outside the trenches and the controlled area: (1) in on-site and off-site surface soils and stream sediments; (2) in soil cores evenly distributed to a depth of 90 cm; (3) in well samples 15-27 m deep; and (4) in a sample associated with evaporator plume discharges.

Primary conclusions which may be drawn from the hydrogeolgical and radiological information include:

- Pu and other radionuclides have been leached or released from the wastes to form a Pu-contaminated leachate.

- There is Pu contaminantion in and around the Maxey Flats burial site.

- Pu has migrated or moved from the trenches and the site possibly via several pathways including; surface transport, interflow, subsurface movement, and atmospheric fallout.

- Samples collected in similar locations as the Pu samples contained H-3, Co-60, Sr-89, Sr-90, Cs-134, and Cs-137, in concentrations greater than background or fallout, indicating that other radioactive contaminants are moving or migrating also; thus tending to confirm the conclusion that Pu is migrating.

- Contamination from other sources such as spills from routine operations, spills during trench dewatering, the use of contaminated fill, and cross-contamination during the sampling has been reported or suggested. After careful review, it has been concluded that the contamination from these other reported sources does not significantly alter the data from the KDHR's "6-month" study or the basic conclusions concerning the potential for migration of Pu via the water pathways.

- There is insufficient radiological and hydrogeological information to estimate in what form, in what quantity, or at what rate Pu is migrating.

Conclusions of a more general nature which can be drawn from experience and data from Maxey Flats follow:

- According to many technical references, Pu normally does not move through the soil and subsurface more than a few centimeters, yet at Maxey Flats, it has been detected tens and hundreds of meters from the trenches.

- The burial site was expected to retain the buried Pu for its hazardous lifetime, but Pu has migrated from the site in less than 10 years.

- If 100% retention of a waste for its hazardous lifetime is the goal of shallow land disposal, continued burial of Pu (and other radionuclides) in humid climates using present waste forms, containers and trench construction methods will not achieve the goal.

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APPENDIX A

CROSS-CONTAMINATION OF SOIL SAMPLES BY USING DIRTY CORING DEVICE

Could significant Pu contamination of the soil samples collected by the KDHR have resulted from cross-contamination by residue left in the coring device from a previous contaminated sample? And if so, what would be the maximum concentrations of Pu transferred into the new sample?

Cross-contamination of a new sample by residues from previous samples certainly is possible if the coring device is not thoroughly cleaned between uses. It is not expected, however, that the amount of residue carried forward from the previous sample would be greater than several thousandths or several hundredths of the total volume of the coring device. If a residue in this amount were carried forward, it would be thoroughly mixed with the new sample during the sample preparation process prior to analysis and would therefore be diluted to several thousandths or hundredths of its original concentration.

Let us take the example of the three cores taken from the same core hole at Trench 26 (see Figure 9). Core No. 139 had a Pu-238 concentration of 4.75 dpm/g; if this contamination resulted from a small amount of residue the left in the coring device, the average concentration of Pu-238 in the previous sample should have ranged from 950 to 240 dpm/g if the new sample were contaminated by a residue carried forward in the coring device equal to 5/1000ths to 2/100ths of the volume of the new sample. Yet if we examine all of the soil cores collected at Maxey Flats, the highest concentration of Pu-238 detected was 20 dpm/g or less than 1/10th of that required to produce the contamination observed in No. 139. Using a similar calculation for Core No. 140, the next deeper core, we can assume that the Pu-238 concentration in the preceeding core was 4.75 dpm/g. If we again assume that 5/1000ths to 2/100ths of Core No. 139 was carried forward to No. 140, the average concentation of Pu-238 contributed by No. 139 to No. 140 would range from 0.024 to 0.095 dpm/g. Yet the concentration found in Core No. 140 was 13.30 dpm/g or almost three times greater than the concentration of Pu-238 found in No. 139. Again, making similar calculations for Core No. 141, the next deeper core from the same core hole, we know that the Pu-238 concentrations in the two preceeding cores were 4.75 and 13.30 dpm/g respectively. Therefore, again assuming that 5/1000ths to 2/100ths of Core No. 140 (13.30 dpm/g) was carried forward to Core No. 141, the average concentration of Pu-238 contributed by No. 140 would range from 0.067 to 0.27 dpm/g. Yet the concentration of Pu-238 found in No. 141 was 4.04 dpm/g. Similar observations can be made about the core samples collected near Trenches 33L, No. 7 and No. 10. Clearly, the dilution

one should expect has not occurred if the contamination in the core samples in the trench area is due to the use of a dirty or contaminated coring device.

It seems clear that generally to the author, the higher surface concentations of Pu-238 which would be required to cause significant contamination of the "6-month" samples were not found at Maxey Flats. And on a detailed basis, such as examining the mechanics and sequence of sampling as at Trench 26, the concentrations of Pu-238 actually are too low to be the source of significant cross-contamination.

APPENDIX B

CROSS-CONTAMINATION OF WELL SAMPLES BY USING DIRTY SAMPLING DEVICE (BAILER)

Could the concentrations of Pu-239 in the samples from the E-Series wells be the result of cross-contamination from a dirty sampler? It is possible that some contamination may have been transferred to a new sample if the bailer used to collect the sample was not thoroughly washed? No, it does not seem probable that all of the Pu detected in the samples collected from the wells was caused by cross-contamination from a dirty bailer for the following reasons:

(1) The concentrations of Pu-238 and ratios of Pu-238/239 in samples from the wells were significantly higher than the average of those found in surface soil and soil core samples collected around the site. It is believed that the sampling process and sample preparation would tend to cause significant dilution of Pu-238 transferred by comtaminated residue from one well into the sample of another well. First, let us make a very conservative assumption that the contaminated residue which was transferred into the sample might make up as much as 10 to 25 percent by volume of the new sample to bring the new sample up to the concentration reported. * If 10 percent contaminated residue were introduced, it would have to have Pu-238 concentrations of 86, 66-75, and 220-330 dpm/g respectively in samples from Wells 3E, 6E, and 11E to raise the Pu-238 concentrations in the samples from the wells up to the levels reported.

It seems more reasonable, in fact, to assume that no more than a trace or several tenths of one precent by volume of contaminated residue would remain in the bailer from a sample collected from a previous well, if any at all remained. In this latter case the contaminant would have had to have a Pu-238 concentration of 4,000-16,000 dpm/g to raise the samples from the wells to the levels reported. Only one of the surface soil and soil core samples had a concentration of Pu-238 greater than 40 dpm/g and only two had concentrations of 20 dpm/g or greater.

* This estimate is based on the following reasoning and using sample 1616 from Well 3E which had a Pu-238 concentration of 8.6 dpm/g: (1) the new sample from the well would make up 75% by volume of the total sample and have a Pu-238 concentration of 0 dpm/g; (2) the contaminated residue would make up 25% by volume of the total sample and would have a Pu-238 concentration of 34.4 dpm/g; (3) the 25% residue and 75% new sample would be thoroughly mixed; and (4) the total sample would come out with a Pu-238 concentration of 8.6 dpm/g.

Therefore, one can reasonably conclude that there is not an obvious place where surface concentrations of Pu-238 were sufficiently high to serve as a source for contaminating the bailer, if the bailer contained an unreasonably high amount of contaminant such as 10 to 25 percent. One could almost say that there is not even a place where surface concentrations of Pu-238 were sufficiently high to serve as a source for contaminating the bailer if the bailer transferred 50, 75, or even 100 percent of the suspended solids forward to the next well.

2. Samples collected from the E-Series wells by NECO and by EPA's RNEB also contained Pu-238 and Pu-239 in concentrations greater than background. The author has assumed that NECO collected their samples from the wells in such a manner as to not cross-contaminate their samples but this has not been discussed with NECO. The author has, however, discussed the sampling techniques used by RNEB and is confident that cross-contamination of RNEB samples is not a significant problem. A detailed description of sampling method used by RNEB will be included in Montgomery's pathways study [9].

3. If we look at all of the analyses which are available for the E-Series wells, we find that for 9 out of 9 wells, the Pu-238/239 ratio was greater than 20; for 8 out of 9, the Pu-238/239 ratio was greater than 25; for 6 out of 9, the Pu-238/239 ratio was greater than 30; the mean or average ratio for 20 samples was approximately 3; the median ratio was 33; and the range was 0.7 to 71.7. Again, the author concludes that there is no well-defined place or area at land surface where the bailer could pick up contamination with Pu-233/239 ratios such as seen in the E-Series wells.

If we look for a surface source area for contamination which has Pu-238/239 ratios similar to the ratios found in the E-Series wells, only at the east end of Trench 26 were ratios above 20 found. In all other cases, the Pu-238/239 ratios of surface soil and soil cores samples were less than 16.4. The Pu-238/239 ratios of 75 percent E-Series well samples were higher than 20. The author is of the opinion that the chances of transferring contaminants from the Trench 26 area individually into each of the wells (3E, 6E, and 11E) are extremely low because there is no obvious or simple way for the bailer to pick up the contamination.

4. The author believes that "the preponderance of evidence" supports the hypothesis that cross-contamination of samples by a dirty bailer did not contribute significantly to the Pu contamination seen in the E-Series wells. At this time, there are 20 analyses of suspended solids from the E-Series wells; of these, 20 out of 20 samples were definitely contaminated with Pu from a source other than fallout. These samples were collected from nine different wells (1E, 2E, 3E, 6E, 8E, 10E, 11E, and 12E) by

three different organizations (NECO, KDHR, and RNEB) and were analyzed by four different labs (AEC HSL, Radiation Management, LFE Labs, and EPA RNEB). The author cannot see how this many organizations and laboratories could have made mistakes with all of the samples collected.

5. Four samples had Pu contamination in the soluble portion of the sample. Cross-contamination of the liquid portion of a sample by a dirty bailer does not seem possible from a materials balance point of view. In collecting the samples, all of the water is poured out of the bailer after each sample is collected from the well. Even if the bailer is not rinsed and wiped dry before the next sample is collected, only a few drops will remain in the bailer to contaminate the next sample. If, for instance, as much as 10 ml would have had to have been 2890 dpm/ml for this amount to raise the concentration of Pu-238 to 28.9 dpm/ml in the sample collected from well 3E if there was no contamination in the well to begin with. And this would require that the 10 ml of highly concentrated contaminated liquid would not be flushed from the bailer when the bailer was immersed in the well water, or else it would be diluted below a point which would give the concentration necessary to yield 28.9 dpm/ml Pu-238. These circumstances seem highly unlikely to the author.

6. The following radionuclides were also found in the E-Series wells: H-3, Co-60, Sr-89, Sr-90, Cs-134, and Cs-137. The presence of these radionuclides tends to support that migration of radionuclides through the subsurface has occurred. Further, tritium is a radionuclide which cannot be transferred by cross-contamination in a dirty bailer in great concentrations because its transfer would be restricted to the water remaining in the bailer and as discussed above no more than 10 ml should be carried forward from one well to the next.

Appendix C. Concentration of Pu-238 and Pu-239 and ratio of Pu-238 to Pu-239 in suspended solids/sediments and soluble portions of water samples collected from test wells at Maxey Flats burial facility

Well Number	Date	Insoluble			Soluble			Laboratory
		Pu-238 (DPM/G)	Pu-239 (DPM/G)	Pu-238/239 (Ratio)	Pu-238 (DPM/L)	Pu-239 (DPM/L)	Pu-238/239 (Ratio)	
1E	4-30-75	6.4±0.4	0.18±0.07	35.6	-0-	-0-	-0-	EPA-RNEB
2E	9-11-75	25.8±1.5	0.6±0.09	43.0	<0.02	<0.01	-0-	EPA-RNEB
3E	2-18-74	8.6±0.7	0.12±0.0009	71.7	-0-	-0-	-0-	AEC-HSL
3E	7-9-74	18.2±1.8	< 0.4	>41.0	28.9±5.1	<2.2	>13	Radiation Management
3E (1)	8-8-74	-0-	-0-	-0-	4.6±0.47	<0.1	>20.9	Teledyne Isotopes
3E	11-8-74	3.2±0.2	0.09±0.02	36.3	6.7±0.9	<0.7	>10.0	LFE
3E	4-30-75	1.3±0.1	0.03±0.015	40.0	<0.15	<0.15	-0-	EPA-RNEB
6E	2-4-74	7.5±0.5	0.22±0.03	34.1	-0-	-0-	-0-	AEC-HSL
6E	5-8-74	6.0±0.6	0.19±0.04	31.6	-0-	-0-	-0-	AEC-HSL
6E (1)	11-74&1-10-75	4.3±1.6	<6.0	>0.7	-0-	-0-	-0-	Radiation Management
6E	8-6-74	-0-	-0-	-0-	<1.1	<0.2	-0-	Teledyne Isotopes
6E	11-8-74	-0-	-0-	-0-	<8	<12	-0-	LFE
6E	4-30-75	0.18±0.04	<0.013	13.3	-0-	-0-	-0-	EPA-RNEB
8E	11-8-74	-0-	-0-	-0-	<0.4	1.7±0.7	<0.3	LFE
8E (1)	4-30-75	33.3±2.0	1.2±0.09	27.8	<0.04	<0.04	-0-	EPA-RNEB
8E	8-15-75	-0-	-0-	-0-	<0.34	<0.34	-0-	EPA-RNEB
10E	9-17-&11-8-74	20.6±1.5	<0.2	46.5	<0.9	3.3±1.6	0.3	LFE
10E	4-30-75	8.2±0.7	<1.3	6.2	<0.15	<0.1	-0-	EPA-RNEB
11E	2-18-74	55±2	0.59±0.07	55.9	-0-	-0-	-0-	AEC-HSL
11E	5-8-74	22±2	0.47±0.07	46.8	-0-	-0-	-0-	AEC-HSL
11E	11-8-74	2.0±0.1	0.07±0.022	30.7	-0-	-0-	-0-	LFE
11E	12-8-74	4.3±1.1	<0.72	>6.0	-0-	-0-	-0-	Radiation Management
11E	4-30-75	0.86±0.09	0.1±0.01	7.8	<0.1	<0.1	-0-	EPA-RNEB
11E	8-15-75	-0-	-0-	-0-	<0.555	<0.555	-0-	Radiation Management
12E	4-30-75	16.2±0.9	0.64±0.13	25.2	<0.06	<0.06	-0-	EPA-RNEB
13E	4-30-75	16.7±0.9	0.75±0.11	22.1	-0-	-0-	-0-	EPA-RNEB

(1) Total sample: insoluble plus soluble.

APPENDIX D

EPA RESPONSES TO THE REVIEW COMMENTS OF THE ENERGY RESEARCH DEVELOPMENT AGENCY ON, "PRELIMINARY DATA ON THE OCCURRENCE OF TRANSURANIUM NUCLIDES IN THE ENVIRONMENT AT THE RADIOACTIVE WASTE BURIAL SITE, MAXEY FLATS, KENTUCKY."

1. The results of studies mentioned on page 11 should be included in the report.

Response: Preliminary drafts of the EPA studies are expected to be available in March. A high priority will be given to their review and they will be issued as soon as possible.

2. A statistically sound basis should be used to distinguish between background radioactivity and that attributable to burial ground operations.

Response: The significance of ERDA's comment is appreciated but is clearly beyond the scope of the present report. EPA will confer with ERDA on what it considers a statistically sound basis for distinguishing between background radioactivity and that attributable to burial ground operations. For the purpose of this paper, concentrations for Pu-238 and Pu-239 were conservatively set well above general regional background levels so that there would be as little question as to whether the Pu detected in a sample represented contamination from fallout or contamination from the wastes buried at the site. It is our opinion that the follow-on papers mentioned in response to question No. 1 above would be the appropriate place for determining what the background levels are. This must be done for a broad spectrum of other radionuclides which have moved or have the potential to move from the site.

3. Some estimate should be provided of the fraction of Pu originally in the wastes which has moved.

Response: Insufficient information is presently available to make the estimate requested. The inventory of radioactivity in the form kept by NECO is inadequate to determine what isotopes, where and how much Pu has been buried at Maxey Flats in any useful form for scientific studies at the site.

It is hoped that the computer-based radioactive waste inventory system which EPA has been developing in cooperation with the State will give us this capability. It may be necessary to investigate individual invoices with the producer of the waste to finally determine the extent of the problem. On the other side, a knowledge of the movement of water

through the hydrogeologic framework at Maxey Flats together with an understanding of how Pu and the other radionuclides buried therein interact with the geologic formations are also required to make the requested calculations. In brief, we don't know what's buried at the site or understand the hydrogeological/geochemical system it is buried in. EPA believes such information is needed and that the studies to obtain this information should be performed.

4. It should be pointed out that each trench and the waste buried therein has a unique inventory of Pu in a unique waste matrix and is therefore a unique source term.

Response: ERDA is correct on this matter and the paper has been modified appropriately. We feel, however, that as these leachates migrate downward into the deeper subsurface formations, they will be subject to mixing, dispersion and dilution within the various perched and regional water bearing formations. The end result may well be the discharge of contaminated groundwater with a somewhat averaged composition but possibly with several subgroups.

5. Some perspective should be provided in the report on the extent to which plutonium movement that has been observed poses a current or possible future hazard.

Response: We can only say that the migration and movement from the site is apparently not causing any health hazard at the present time. Insufficient information is available to make an estimate of the situation which may exist in the future.

6. The term "migration" is variously used in the paper to describe different mechanisms of movement. To avoid the resultant ambiguity and possible misunderstanding, the term should be used as it usually is in this field, i.e., to mean movement through soil. It should not be used to mean all mechanisms of transport.

Response: The use of "migration" throughout the paper has been reviewed and changed so far as practical without changing the meaning or intent of the paper.

7. The statement on the top of page 24 should not be underlined. Underscoring the possibility that "...Pu may be migrating from the trenches via the subsurface pathway" (page 24) adds emphasis that is not warranted by the data presented in the paper.

Response: The underline has been removed.

8. Units (dpm/g) and estimates of error should be given in Table 1.

Response: These editorial comments were rejected. The reader can make a rough estimate of the error of analysis for the soil and soil core samples by looking at the estimated error for the suspended solids analyses in Appendix C.

9. Surface and subsurface sample data should be presented separately to avoid misunderstanding and misinterpretation.

Response: These editorial comments were rejected. The data are separated for interpretation purposes in Table 1 into what the author considers surface samples (i.e., ON-SITE and OFF-SITE) and subsurface samples (i.e., SOIL CORE and TEST WELL).

10. Probable errors in Pu-238/Pu-239 ratios should be reported.

Response: There are several methods for calculating the probable error for a ratio of two values each of which has its own estimated error of analysis. However, it is the author's opinion that no one of these methods is satisfactory to all statisticians or for all purposes. Therefore, the author prefers to furnish the estimated error of analysis for the basic data and let the reader determine the probable errors in the Pu-238/Pu-239 ratios as he wishes.

11. An estimate should be provided of the amount of Pu in the large mobile source term mentioned on page 30. If some quantitative estimate cannot be inferred from the data, the statement should be deleted.

Response: There is insufficient information on the source term of Pu buried at Maxey Flats. However, based on a preliminary review of the computer-based inventory data, the total source term of Pu-238 and Pu-239 might run as high as 200-300 kg. As a bare minimum, we know that 80 kg Pu-239 have been buried at the site. This is a large source term; however, the mobile fraction is unknown.

12. Beginning on page 25 in section 7, the discussion of information presented in reference 34-42 should be improved to better represent the findings of the authors.

Response: The discussion reflects the author's understanding of the references.

13. The projected burial growth rates for volumes of wastes and quantities of special nuclear material, indicated by the arrows in Figure 5, page 35, are at odds with the plotted data, which show a steady decrease since 1972. The arrows should be eliminated or modified appropriately.

Response: Both the text and the title of Figure 5 state that projections shown were based on the information available as of 1972, the period of time when authorities from the KDHR became concerned about the water in the trenches and the phenomenal growth of waste volume and special nuclear material being buried at Maxey Flats. The author showed all of the detail available as a service to the reader.

14. If any additional data on Pu analyses on Maxey Flats environmental samples is available and pertinent, it should be added to the report.

Response: Additional Pu analyses which are available will be published in the studies mentioned in response to comment No. 1.

15. References to documents which are not readily available, particularly personal communications, should be eliminated.

Response: The intent is to indicate the source of additional relevant information. In some cases, personal communications were the only source of information.

16. According to Figure 7, sample 124 was an off-site sample; therefore, the data for sample 124 in Table 1 should be listed with the off-site samples. Also, the Pu-238/Pu-239 ratio for this sample should be corrected.

Response: The Pu-238/Pu-239 ratio for sample No. 124 has been corrected. In regards to ERDA's comment concerning No. 124 being an off-site sample, it falls well within the basic off-site sample description of, "close in to the buried area."

17. As is recognized in the table and elsewhere in the text, the paper presents only preliminary data. It is essential, therefore, that a clear distinction be made between results, conclusions based on results, and speculations offered in the absence of definitive information. In this regard, a number of conclusions are misleading or are not supported by the data presented in the paper. The following are examples:

A. The conclusion on page 31 regarding confirmation of Pu movement provided by measurements of other radionuclides is not supported.

Response (17A): The author feels that finding the other radionuclides in the monitoring wells does tend to confirm the migration of Pu, since these others act as multiple tracers.

B. The data indicate that Pu has been detected some distance from the trenches. However, these preliminary data do not warrant the implication that the many technical references which conclude that Pu does not move very far "through the soil and subsurface" are incorrect. Therefore, the observation that Pu is not supposed to move (migrate) etc., should be removed from the conclusions.

Response (17B): It is the feeling of many authorities that plutonium does not "move" very far through the soil and subsurface rocks. In this case, however, plutonium was found in 20 out of 20 samples available from nine wells and in 14 out of 14 soil cores. If the hypotheses presented in this paper to explain the movement of plutonium are correct, then under certain sets of conditions, the statement needs to be amplified to recognize that plutonium may not be held by the soil and rocks under all possible conditions. We suggest, therefore, that a more careful statement needs to be made about migration through the soil than has been the case until today.

C. The last conclusion on page 31 should be deleted since it is not supported by the limited nature of the survey and the resulting preliminary data.

Response (17C): Additional data has been added to the text which the author feels supports this conclusion.

D. The conclusion on page 31 beginning "Other modes of contamination..." should state which of the water pathways are supported by the data in the text.

Response (17D): This statement has been modified.

APPENDIX I

EPA RESPONSES TO THE REVIEW COMMENTS OF THE NUCLEAR REGULATORY COMMISSION ON, "PRELIMINARY DATA ON THE OCCURRENCE OF TRANS-URANIUM NUCLIDES IN THE ENVIRONMENT AT THE RADIOACTIVE WASTE BURIAL SITE, MAXEY FLATS, KENTUCKY."

1. The paper fails to give adequate emphasis to the public health significance of the data and conclusions presented. Public health significance is referenced in only two sentences on pages 1 and 4.

Response: The fact that health authorities and scientists from the State of Kentucky, the Nuclear Regulatory Commission, and EPA have all advised that the levels of plutonium detected in the environment at Maxey Flats do not constitute a health hazard at this time has been given prominent coverage in both the Foreword and in the text.

The immediate public health significance of the possible migration of Pu at Maxey was not relevant to the point of the paper. The main issues in the paper are that: (1) Pu and other radionuclides are out of the trenches and (2) Pu may be migrating via the water pathways.

2. The paper does not give recognition to the fact that the Maxey Flats burial ground has been under a program of continuing evaluation since the time it was licensed.

Response: We recognize that the regulatory agencies, i.e., the State of Kentucky and NRC have had a continuing interest. The report itself is based on a specific period of time and did not attempt to evaluate the regulatory aspects of waste management, which is properly the responsibility of those regulatory agencies.

3. The paper is based on data collected by Kentucky during the period 11/73 to 5/74. There is little or no new data presented in the paper. Kentucky has been collecting and analyzing samples since conclusion of the 6-month study in May of 1974. Also, the EPA and USGS have been conducting research studies of the site. Preliminary results of these studies and longer term environmental monitoring data may support or deny certain of the paper's conclusions.

Response: We recognize the paper is limited in time, and remedial improvements made since May of 1974 have been recognized. Completion of preliminary drafts of EPA, USGS, and KDHR site investigations is expected in March 1976. Making these studies publicly available has

been given a high priority. They will be forwarded to NECO, NRC, ERDA, and KDHR, for comment as soon as the drafts are available and will be published as soon as their review is completed. EPA plans to keep the review time short and issue these reports as soon as practicable because of the widespread interest in their content.

4. The first three objectives of the paper have already been accomplished and presented in the Kentucky 6-month report. Also, it appears desirable to attempt the fourth objective using only six months of data, when, as indicated in the paper, additional data is available.

Response: Delay of the paper until all of the facts and all of the studies were completed was, of course, possible. However, it is believed that presentation of the paper at San Francisco at a symposium convened specifically to present new findings on transuranium nuclides in the environment was particularly appropriate. Now that the paper is common knowledge and will be publicly available in the proceedings of the symposium, EPA cannot justify withholding its release. The interpretations presented in the paper were presented for public review, comment, and criticism.

5. As the title suggests, the paper is preliminary in nature since it presents several specific and general conclusions concerning pathways for the migration of plutonium based on data which the author concedes equally support or do not rule out other "possibilities." We believe that before predicting the future effectiveness of Maxey Flats burial ground, the degree to which site operations and each pathway is contributing to the radioactivity detected off-site should be determined.

Response: We concur with the need for additional information on pathways contributing radioactivity detected off-site. However, the extended period of time that may be required to gather this information makes it impracticable to include such information in this edition. We still believe that the information in the existing report will be of interest to those concerned with the problem of possible plutonium migration through the soil. In answer to the entire problem of shallow land burial, site operations will be addressed on a continuing basis as the information is gathered by EPA and the other agencies.

6. The paper speaks of plutonium migration by any of four modes; (1) surface runoff, (2) interflow, (3) subsurface interflow, and (4) atmospheric dispersion. We believe use of the term migration should be limited to discussion of the movement of the material through the soil (interflow) and underlying geologic formations (subsurface flow). Surface runoff carrying contamination from site operations and evaporator fallout are contributing radioactivity to the local environment which makes it difficult to determine to what extent radioactivity has migrated or is continuing to migrate via the interflow and subsurface flow modes.

Response: The use of "migration" throughout the paper has been reviewed and changed so far as practicable.

General Comment: EPA may want to consider a second overview paper which takes into consideration the shortcomings of this paper. Such a paper could summarize the current status of the various evaluation activities being conducted at the site, present longer term trend data, current monitoring data, discuss the public health significance of the data, evaluate the effect of the improved water management program and discuss the critical pathways contributing radioactivity to the local environment in terms of the recent data. We believe such an overview summary could provide a useful reference document for those agencies and parties having responsibilities while still serving to focus attention on a situation that merits continued monitoring and study.

Response: EPA already plans to publish additional reports concerning the Maxey Flats burial site. These reports will present longer term trend data, current monitoring data, discuss the potential public health significance, hydrogeology, and discuss the critical pathways contributing radioactivity to the local environment. We do not, however, have any information on the effects of the improved water management program.

General Comment: The paper's closing conclusion notes that the goal of waste disposal sites, i.e., 100 percent retention of wastes for their hazardous lifetime, is not being achieved. It is our opinion that 100 percent retention is not a realistic goal. It is doubtful that a site could be found that would not permit some releases albeit small. Disposal sites should be selected and operated in such a way as to minimize releases to off-site areas. We agree that criteria for evaluating releases of radioactivity from commercial burial grounds should be defined to assist in determining the acceptability of existing and future disposal sites and disposal practices.

Response: EPA's underlying philosophy is that waste management means containment of radioactive materials until they have decayed to innocous levels. The objective is to minimize exposure to present and future populations and to avoid dilution into the biosphere. Containment may involve burial, storage, or some other form of assuring that dispersion into the biosphere does not take place.

EPA believes this philosophy is consistent with the criteria under which burial sites, such as Maxey Flats, were initially licensed. This criteria was spelled out in the draft environmental impact statement for the Liquid Metal Fast Breeder Reactor (WASH-1535) as "The basic objective of shallow land burial is the confinement of radioactive materials on the site over the periods of time the wastes remain a hazard. Site evaluation and field studies should provide reasonable assurances that this objective will be met."(1) In addition, the same criteria can be found in the draft Generic Environmental Statement Mixed Oxide Fuel (WASH-1337) and the Draft Environmental Statement for the Uranium Fuel Cycle (WASH-1248).

The Task Force on Radioactive Waste Management of the Conference of State Radiation Control Program Directors, in its 1974 Annual Report to the Conference stated: "Authorization to operate a commercial land burial facility is based on an analysis of the nature and location of potentially affected facilities; of the site topographical, geographical, meteorological, and hydrological characteristics; and of groundwater and surface water use in the general area which must demonstrate that buried radioactive waste will not migrate from the site."(2)

EPA believes that the development and verification of environmentally acceptable techniques for disposal and long-term containment of low-level radioactive wastes is a significant problem that needs to be solved soon. EPA is optimistic that this problem can be solved. However, a number of actions may be required to achieve this goal including: development of criteria for selection and evaluation of sites; limiting the quantities of certain radionuclides allowed to be buried, based on half lives and relative biological hazards; segregation of waste at the source; new packaging or containerization requirements; more selective choice of sites; and additional operational controls at the sites themselves.

EPA intends to pursue a program to develop minimal acceptability criteria in terms of environmental barriers, and also to address the problems related to monitoring around sites, and the development of action levels that may be necessary to protect man and his environment. EPA intends to carry out this program in cooperation with the other agencies involved.